

Atmospheric System Research (ASR) Science and Program Plan

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Executive Summary

Anthropogenic increases in atmospheric greenhouse gases, such as carbon dioxide, and in aerosol particulate matter are impacting the current climate and will continue to influence the future climate. Climate models must understand and represent the extent of these influences on the current climate, as well as the prospective influences of past, current, and future emissions on the future climate. Current climate models show a large spread in the values of projected climate parameters like surface temperature and precipitation, and this spread makes it difficult for policy makers to use such projections to develop national and international energy policy. The spread in climate model projections arises from two broad sources of uncertainty. First, scientists believe that anthropogenic *atmospheric aerosols* partially offset the global warming influence due to enhanced greenhouse gas concentrations, but because climate models are uncertain how to represent the complex aerosol lifecycle in the atmosphere, they are also uncertain about the impact of aerosols on climate. Second, *clouds* are a large source of uncertainty in climate models, particularly how clouds will respond to and interact with changes in atmospheric composition. Both aerosols and clouds influence *radiation and precipitation*, which together largely drive the global atmospheric circulation.

In order to improve the fidelity and predictive capability of global climate models, a better understanding of various fundamental aerosol and cloud life-cycle processes is required. Furthermore, this understanding must lead to more accurate model representations of these processes and their interactions with the radiative, dynamic, and thermodynamic properties of the atmosphere. To achieve these high-priority climate research objectives, modelers and observers must develop and apply comprehensive data sets to constrain and guide model development and evaluation.

The Department of Energy (DOE) is uniquely positioned to address these challenges. DOE has been an active participant in climate research for decades, as energy production is one of the largest sources of anthropogenic greenhouse gases and aerosols, and future energy production and policies will be dependent upon climate conditions. As such, DOE has the modeling and observational expertise to address the uncertainties in the atmospheric processes that currently hinder climate models. This plan describes a new observation-based DOE research program that will systematically improve the understanding and model representation of aerosol, cloud, and precipitation processes and properties.

The DOE Atmospheric System Research (ASR) program, which combines the capabilities of prior DOE observation-based programs, will advance process-level understanding of the key interactions among aerosols, clouds, precipitation, radiation, dynamics, and thermodynamics, with the ultimate goal of reducing the uncertainty in global and regional climate simulations and projections. To accomplish this goal, the ASR program will work to address the following process-related research objectives:

- Determine the properties of and interactions among aerosols, clouds, precipitation, and radiation that are most critical to understand in order to improve their representation in climate models.
- Ascertain the roles of atmospheric dynamics, thermodynamic structure, radiation, surface properties, and chemical and microphysical processes in the life cycles of aerosols and clouds, and develop and evaluate models of these processes.

- Identify and quantify processes along the aerosol-cloud-precipitation continuum that affect the radiative fluxes at the surface and top of the atmosphere and the radiative and latent heating rate profiles, and improve the ability to accurately model these processes.

As an observation-based research program, ASR will be tightly coupled with the Atmospheric Radiation Measurement (ARM) Climate Research Facility. The ARM Facility is a DOE national user facility with extensive long-term ground-based in situ and remote sensing observations that are supplemented with episodic airborne observations and laboratory studies. The fixed ARM sites are situated in climatically distinct locations to sample tropical, mid-latitude, and Arctic environments, and mobile facilities are used to sample other climatically important locations. ARM's observational capability has been greatly increased by the recent American Recovery and Reinvestment Act, allowing better observation of many of the processes along the aerosol-cloud-precipitation continuum. The automated, long-term ARM observations will uniquely facilitate the analysis of the various modes of variability such as diurnal, synoptic, and seasonal variation in the cloud and aerosol life cycles. ASR scientists will use the ARM observations and process-level models to improve process-level understanding by concentrating on the following approaches:

- Determine the essential characteristics of the coordinated laboratory and field measurements necessary to understand aerosol and cloud life cycles and aerosol-cloud-precipitation-radiation interactions (e.g., variables measured, measurement accuracy, sampling strategies).
- Develop research strategies to create the integrated data products necessary to improve the understanding of aerosol-cloud-precipitation-radiation interactions (e.g., retrieval development, uncertainty analysis, data product collocation, quality control).
- Utilize the integrated data products to develop, evaluate, and ultimately improve the parameterization of aerosol-cloud-precipitation-radiation processes in models over a range of scales (e.g., what data products are needed, what metrics are used to evaluate model improvement).
- Evaluate the tradeoff between the minimum level of model complexity and model accuracy required to represent the range of atmospheric conditions that determine climatically relevant aerosol and cloud properties and aerosol-cloud-precipitation-radiation interactions.

Combining ARM's observational capability with the ASR scientific expertise will result in a better understanding of atmospheric processes that comprise the cloud and aerosol life cycles, and the interactions among them, all of which are essential to improve the accuracy of climate models. Thus, ASR will support the development of national energy and climate policy by achieving improvements in the understanding and modeling of climate system components that have been identified as major sources of uncertainty in climate projections.

Acknowledgments

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1.0 Introduction

Increases in the concentrations of atmospheric greenhouse gases and aerosol particulate matter that result from human activities, and in particular from energy production, are impacting current climate and may be expected to exert an even greater influence on future climate. These anthropogenic influences and the climate system response are well documented by recent reports from the Intergovernmental Panel on Climate Change (IPCC 2007a,b), the authoritative international body charged with reviewing the global understanding of climate and climate change. However, the large spread in projected future surface temperatures from current global climate models for any given assumed future anthropogenic emission scenario greatly limits the usefulness of these models in the development of the nation's and the world's energy policies.

One of the leading sources of uncertainty in global climate models is the radiative influence (forcing¹) of aerosols, which is believed to be offsetting a substantial fraction of the warming influence due to enhanced greenhouse gas concentrations. The sensitivity of the climate system response (feedback²) to this radiative forcing is another leading source of uncertainty, due in large part to unknown subsequent changes in the amount and properties of clouds that may augment or diminish the effects of the initial radiative forcing. Difficulties in representing forcings and feedbacks in global climate models translate into a large uncertainty in the ability to predict trends in the global mean surface temperature and other changes in climate that would result from a specified increase in the amount of atmospheric carbon dioxide.

Among the agencies of the U.S. Government working to understand the impacts of human activities on the Earth's climate and their implications on society, the U.S. Department of Energy (DOE), as the steward for U.S. energy policy, has long had principal responsibility for understanding how energy production may result in climate change and how climate change may impact energy production. DOE has been a key member of the multiagency U.S. Global Change Research Program (USGCRP) since its inception. During the past three decades, the Office of Biological and Environmental Research (BER) in the DOE Office of Science has sponsored programs that have focused on specific aspects of climate models to improve the understanding of processes that are represented in these models. These research programs have consisted mainly of long- and short-term observation-based field investigations, laboratory studies, numerical modeling efforts, and theoretical work. Together, these programs have contributed to major improvements in the representation of physical processes in climate models; however, these efforts have also demonstrated that much more work remains to be done in order to achieve the predictive capability of climate models that is needed for policy-making purposes.

The principal observation-based DOE programs examining atmospheric processes that must be understood and represented in climate models have been the Atmospheric Radiation Measurement Program (ARM Program), the Atmospheric Science Program (ASP), and the ARM Climate Research Facility (ARM Facility). The ARM Facility was established by BER to collect long-term ground-based and in situ data in several climatic regions around the world; it has now been designated a national user facility. Data from ARM sites are used to support a wide range of international atmospheric and climate

¹ "Radiative forcing" is a measure of the influence a factor has on altering the balance of incoming and outgoing radiant energy in the Earth-atmosphere system.

² A "feedback" is an interaction mechanism between processes in a system, where the result of a change in an initial process triggers changes in a second process, which in turn influences the initial process again.

research. The passage of the American Recovery and Reinvestment Act (ARRA) in 2009 provided an opportunity for BER to dramatically expand the observational capabilities of the ARM Facility. These new observational tools, together with the instrumentation already in use (Appendix A), will allow better characterization and quantification of climatically important atmospheric processes. The enhanced ARM Facility instrumentation will provide data sets that allow for evaluation and improvement of various components of climate models that have previously been difficult to assess. To better utilize these new tools, BER has merged the ARM Program and ASP into a cohesive unified program called Atmospheric System Research (ASR). This document outlines the programmatic objectives of ASR and serves as the science plan for this program.

1.1 Historical Background

For the past two decades the ARM Program and ASP have been the two research programs within the BER's Climate and Environmental Science Division (CESD) charged with conducting research on atmospheric processes pertinent to climate and climate change. These programs have taken coordinated and complementary approaches to quantify the effects of clouds and aerosols on the atmosphere's radiation balance. Both programs have used laboratory studies, atmospheric measurements, and numerical modeling to examine processes that influence atmospheric radiative transfer in order to increase the fidelity of process representations in climate models. These programs have supported the development and use of models on various scales, with domains that span the range from box models (order of meters) to global, to evaluate and improve the ability to make climate projections for assumed emissions scenarios. Thus these programs have provided data sets needed for the development of climate models in the United States and internationally and have played a direct role in these model development activities. The objectives, approaches, and key accomplishments of the two programs are briefly presented here; examples of recent research findings of the two programs are presented in Appendix B.

The ARM Program, which was begun in 1990, has greatly advanced the treatment of cloud and radiative processes in climate models (Ackerman and Stokes 2003). The original programmatic objectives of ARM (Stokes and Schwartz 1994) were to:

1. Relate observed radiative fluxes and radiances in the atmosphere, spectrally resolved and as a function of position and time, to the temperature and composition of the atmosphere, specifically including water vapor and clouds, and to surface properties, and to sample a sufficient variety of situations as to span a wide range of climatologically relevant possibilities.
2. Develop and test parameterizations that can be used to accurately model radiative influences of water vapor and clouds, with the objective of incorporating these parameterizations into global climate models (GCMs).

To address these goals, the ARM Program was initiated to gather long-term, collocated, ground-based data sets on radiation, clouds, and atmospheric state. During the early years of the program, ARM focused on site development and, in particular, the development of advanced instrumentation that could be operated autonomously and over long time periods at the ARM Facility sites. The program fostered the successful transition of several instruments that were previously manually intensive into truly operational instruments; these instruments include state-of-the-art sensors such as millimeter-wavelength cloud radars, high spectral resolution infrared Fourier transform spectrometers, micropulse cloud lidars, and water vapor Raman lidars. These advanced sensors, together with more traditional observing tools

such as multiple radiosonde ascents per day, passive microwave radiometers, broadband hemispheric radiometers, surface meteorological observations, and sky imagers, are the core observational tools at each ARM Facility site.

To collect data across a wide range of conditions, the ARM Program established fixed sites in locales characteristic of different climatic regimes (DOE 1991). These sites (Figure 1) are in the mid-latitudes (the extensive Southern Great Plains site in Oklahoma, established in 1992), the Arctic (the North Slope of Alaska site in Barrow, Alaska, established in 1997), and in the tropical Pacific Ocean (the three Tropical Western Pacific sites established in Manus, Papua New Guinea in 1996; on Nauru Island in 1998; and in Darwin, Australia in 2002). To sample additional climatic regimes, the program developed the ARM Mobile Facility (AMF), which became operational in 2005 and which has been deployed in several campaigns. A second mobile facility, currently under construction, will become operational in 2010. In addition to the fixed sites and the mobile facility, the ARM Facility also maintains an airborne facility for in situ sampling of clouds, aerosols, and radiation. In 2004, DOE declared the ARM observational sites to be the ARM Climate Research Facility, making the ARM Facility a formal national user facility. ARM provides support to the scientific community through experiments funded via peer-reviewed proposals.

The development of the ARM Facility's observational tools, especially the automation of advanced ground-based remote sensors, is a significant accomplishment of the ARM Program. Using these tools, the ARM Program has made considerable progress towards its original programmatic objectives. Key accomplishments are improvement in the accuracy of longwave and shortwave radiative transfer models (now better than 1% under clear sky conditions), the compilation of detailed vertically resolved cloud climatologies at several sites, marked improvement in retrieving cloud properties from active and passive remote sensors, important advances in measuring the radiative impact of various types of clouds both at the surface and within the atmosphere, the first ground-based quantified observations of the aerosol-cloud albedo effect, the development of an objective approach to use ground-based observations to drive single-column and cloud resolving models, and numerous advances into understanding the microphysical processes in liquid, ice, and mixed-phase clouds. Some scientific highlights from the ARM Program over the previous five years are presented in Appendix B.

DOE has a long history of atmospheric chemistry and aerosols research. The ASP and its predecessor programs contributed to a better understanding of gas-phase oxidation chemistry that is vital to understanding secondary organic aerosol formation and particle aging, which are processes that continue to be important avenues of research today. Additionally, DOE focused effort on understanding aqueous-phase chemical processes within cloud droplets and wet removal processes, which are mechanisms important to aerosol processing by clouds, and dry deposition and resuspension of aerosol particles pertinent to the amount and properties of atmospheric aerosols.

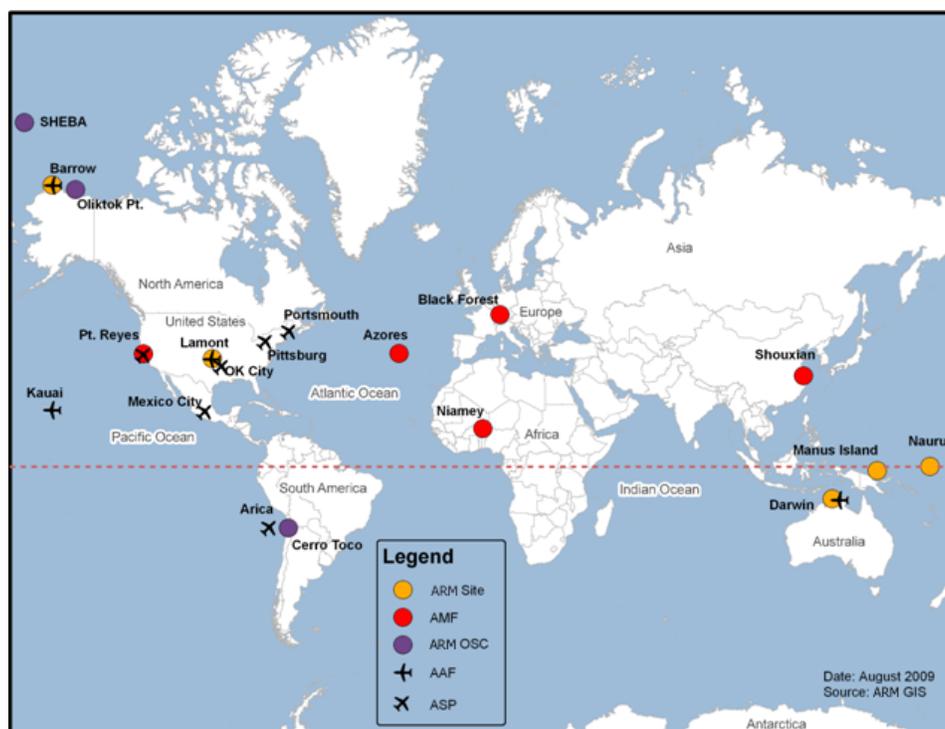


Figure 1. The locations of the fixed ARM Facility sites, the previous and current ARM Mobile Facility (AMF) deployments, the short-term off-site ARM campaigns (OSC), and the ARM Aerial Facility (AAF) and ASP aircraft experiments since 2003.

Recognizing the outstanding uncertainties in the climate impacts of atmospheric aerosols, DOE reconfigured the ASP in 2004 to concentrate primarily on these aerosols and their climate influence (BERAC 2004). In particular, the ASP focused on the processes that govern the properties of natural and anthropogenic atmospheric aerosols that most influence climate and climate change: the amount, spatial distribution, composition, size distribution, optical properties, and cloud nucleating properties. ASP research objectives were to improve understanding and models for:

1. Aerosol life cycle, especially new particle formation, growth, and secondary aerosol formation (especially secondary organic aerosol), and including the effects of primary particle emissions, precursor gases, and cloud processing; the influence of these processes on amount- and size-distributed composition of aerosols; and the modification of aerosol amount and properties by anthropogenic emissions.
2. Aerosol optical properties, their dependence on relative humidity, and the relation to size-distributed composition.
3. Aerosol cloud nucleating properties, including the influences of aerosols on cloud microphysical properties.

A major element of ASP research consisted of field measurements of the climate-relevant processes and properties of different aerosol types. Field campaigns are the key to both quantifying aerosol properties and atmospheric processes under a variety of locations and atmospheric conditions and testing and improving the accuracy of models.

Like ARM, ASP supported an instrument development component. This activity achieved major advances in particle size and composition measurement through single-particle mass spectroscopy, pioneering instrumentation for the detection and composition analysis of particles having diameter less than 3 nm, thereby nearly bridging the gap between molecular cluster precursors and freshly nucleated aerosol particles, and fast-response instrumentation for rapid measurement of aerosol size distributions and selected gaseous precursors to achieve good spatial resolution during aircraft sampling. These techniques have been deployed in the field to great advantage. Recent field campaigns have provided in situ measurements of cloud drop number and size distribution in marine stratocumulus clouds, which in conjunction with gust-probe measurements of vertical velocity, are proving to be invaluable for testing theories and parameterizations of cloud droplet and drizzle formation.

In addition to field measurements ASP research encompasses laboratory studies of reactions of aerosol precursor gases and of interactions between aerosol particles and reactive gases, laboratory measurements of aerosol optical properties, process-level modeling, and regional modeling in support of the field campaigns. Some key highlights that have resulted from this work over the past five years are presented in Appendix B.

The ARM Program and ASP were both observation-based programs and had several common interests in certain atmospheric processes (e.g., aerosol-cloud interactions) and related goals (e.g., improving process representation in global climate models), but the research approaches of the two programs emphasized different measurement scales. ARM science studies have been based mainly on the long-term observational record from the heavily instrumented ARM Facility fixed sites and the ARM Mobile Facility, supplemented with periodic intensive field campaigns at these sites. In contrast ASP research has made extensive use of controlled laboratory experiments to study key atmospheric processes, especially those involving chemical reactions or composition-dependent aerosol properties, and extended the studies of these processes into the atmospheric environment using in situ measurements in intensive short-term field campaigns. In the past several years, the two programs have coordinated several joint field campaigns, typically at ARM Facility sites, thereby combining the distinct measurement scales, methodologies, and expertise available within the two programs to yield complementary data on the same processes, which has led to new insights and understanding.

The new ASR program will continue to rely heavily on measurements from the ARM Facility. Through funds provided under the America's Recovery and Reinvestment Act of 2009, the ARM Facility is acquiring extensive new measurement capabilities, including scanning cloud radars, precipitation radars, additional aerosol observations, flux measurements for its surface sites, and enhanced aerosol and cloud measurement instrumentation for its aerial facility (See Appendix A for more details). These new capabilities will allow the combined program to extend its research horizon, especially in cloud dynamics and precipitation studies, and work towards the integrated vision described in the following sections.

1.2 Scientific Motivation

The primary objective of climate research in the United States and internationally is to develop understanding of the processes that control climate change and to represent this understanding in models, specifically general circulation models (GCMs), which are often referred to as global climate models. GCMs are detailed models that aim to represent the full range of climatically relevant physical processes within the atmosphere and the oceans. These models are the most powerful tools available to study the

climate impacts of alternative future emission scenarios. However, there is substantial divergence at present among state-of-the-art models in their sensitivities (i.e., the amount by which global mean temperature changes for a given change in atmospheric composition) that limits the confidence that can be placed in projections of future climate change obtained with these models.

The latest assessment report by the Intergovernmental Panel on Climate Change (IPCC 2007 a,b) has shown that despite the ability of multiple GCMs to accurately reproduce the increase in global temperature over the twentieth century, there is considerable uncertainty in the predictive capabilities of GCMs. This is evidenced by the large differences of global mean surface temperature as projected by multiple models for each of several assumed future emission scenarios (Figure 2). The spread among the models can be attributed, in large part, to uncertainties in how changes in cloud amount and properties affect the global radiative budget and the vertical distribution of the radiant energy in the earth-atmosphere system. Reducing the uncertainties in GCM projections requires that the treatment of clouds, and their impact on the planetary energy budget, be substantially improved.

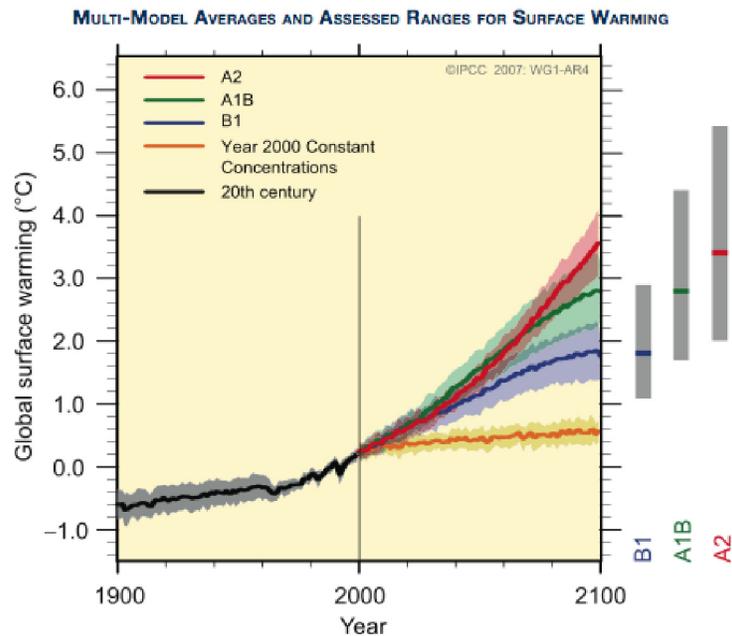


Figure 2. The solid lines are multi-GCM global averages of surface warming (relative to 1980-1999) for various emission scenarios, shown as continuations of the 20th century simulations. The orange line is the experiment where carbon dioxide levels were held constant at year 2000 values. The shading around these solid lines represents the ± 1 standard deviation of the individual model annual averages. The gray bars on the right indicate the best estimate (solid line) and likely range associated with the various scenarios. These gray error bars include both the spread of the mean results from different GCMs, the uncertainties within each GCM that was used in the simulation, and results from a hierarchy of independent models and observational constraints. Modified from IPCC (2007a).

Earth's energy budget, illustrated schematically in Figure 3, is complex and is controlled by a variety of processes that the ASR program will examine. Incoming solar energy is absorbed in the atmosphere by water vapor, clouds, and (to a much lesser extent) aerosols; absorbed by the Earth's surface; and reflected back to space. Infrared energy is emitted by the Earth's surface. This thermal infrared (or longwave)

radiation is largely absorbed by various components in the atmosphere (clouds, water vapor, and trace infrared active greenhouse gases, such as carbon dioxide, methane, nitrous oxide, chlorofluorocarbons, and ozone); these substances in turn emit longwave radiation both back towards the Earth, contributing to the energy taken up at the surface, and upward to space. The infrared radiative flux emitted into space is substantially less than that emitted at the surface due to the lower temperature in the atmosphere as compared to the surface temperature, which is the essential manifestation of the greenhouse effect. On the global and annual average, to a very good approximation, the outgoing infrared energy balances the absorbed incoming solar energy. However, numerous processes modulate the flow of shortwave and longwave radiative energy through the climate system. Clouds, which are highly variable in altitude, space, and time, exert a major influence on radiative fluxes by scattering and absorbing solar and infrared radiant energy. The impact of clouds on the radiative field depends on both the macrophysical (e.g., height, extent) and microphysical (e.g., phase, cloud particle size) properties of the clouds. Aerosols play a secondary but important role in radiative energy distribution by both scattering and absorbing shortwave radiation, and, very importantly, by influencing cloud macrophysical and microphysical properties.

The overall radiative impact of aerosols, both directly and indirectly, depends on the amount of aerosol and its chemical and microphysical properties, and on environmental conditions (e.g., relative humidity and amount of sunlight, reflectivity of underlying surface or clouds). All of these processes and their effects on radiation and the thermodynamic environment must be understood and represented in climate models. The several fluxes shown in Figure 3 are highly variable in space and time, and even in global and annual average are much more uncertain than indicated by the precision of the numerical values given in the figure; understanding the controlling processes and representing them in models to reduce the uncertainties in these quantities is a major objective of the research conducted by the ASR program.

Figure 3 presents the context in which the influences of increases in concentrations of greenhouse gases and aerosols over the industrial period and prospective future increases must be considered. Increases in greenhouse gas concentrations over the industrial period have contributed about $3 \pm 0.5 \text{ W/m}^2$ to the approximately 333 W/m^2 of back radiation shown in Figure 3, or about 1% of the natural greenhouse effect; this is the primary cause of anthropogenic climate change. Anthropogenic aerosols are estimated to contribute about $1.5 \pm 1.0 \text{ W/m}^2$ to the reflected solar radiation, again about 1% of the total. This enhancement of reflected solar radiation tends to offset the warming influence of incremental greenhouse gases. Understanding and quantifying these perturbations in Earth's radiation budget over the industrial period and the climate response to these perturbations is the challenge that faces the climate research community, including ASR.

Uncertainties in GCM projections of the increase in global mean temperature for a prescribed increase in carbon dioxide (which contribute to the gray error bars on the right side of Figure 2) are quite large, and have not appreciably changed from values derived two decades ago. The main factor contributing to this model uncertainty is the impact of clouds and in particular how clouds will evolve and further impact the energy budget in a changing climate (i.e., cloud feedbacks on the model climate). These processes are not well understood, and it has been recognized since the late 1980s that the equilibrium sensitivity of climate models (the change in global temperature that would result from a given sustained radiative perturbation) is highly sensitive to alternative treatments of cloud processes (Cess et al. 1989). It is this situation that motivated the formation of the ARM Program in 1990. The formation of clouds is determined by air motions at many scales, from local to the general circulation. Clouds, through surface and atmospheric radiative heating and through latent heating associated with precipitation, influence the ambient environment and the atmospheric motion on a variety of scales. Thus, atmospheric motions and clouds

are intimately coupled. Furthermore inhomogeneities in the atmosphere (e.g., water vapor, vertical velocity) and aerosol properties impact the clouds in important ways and can influence cloud evolution, leading to potentially important impacts on atmospheric motion.

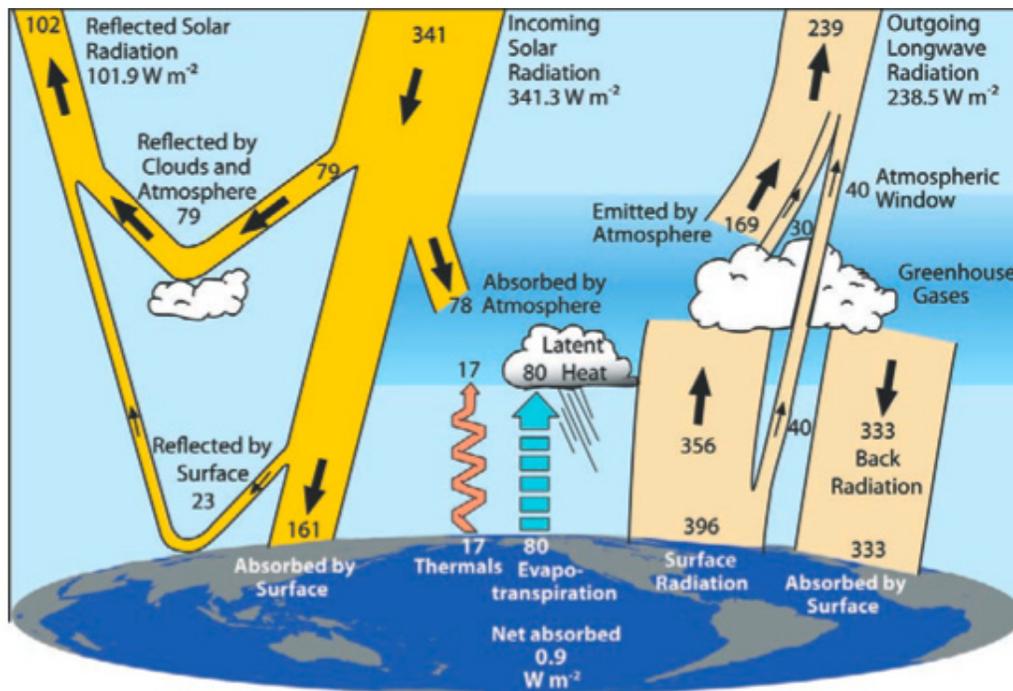


Figure 3. The components of the global mean energy budget of Earth for March 2000 to May 2004, in W/m^2 (Trenberth et al. 2009). The broad arrows indicate the schematic flow of energy in proportion to their magnitude. Several of these fluxes are not as well known as indicated by the precision of the numerical values presented. The net energy imbalance of 0.9 W/m^2 is not considered well established from either satellite measurements or assessments of the increased planetary heat content.

Precipitation plays a very important role in both the energy and hydrological cycles of the atmosphere. The energy balance of the global atmosphere is a balance between the net radiative cooling of the atmosphere and the latent heating of the atmosphere associated with precipitation. Cloud feedbacks in climate model simulations not only affect the radiative heating of the atmosphere, but also determine the response of the hydrological cycle to a changing climate. Furthermore, changes in precipitation location and amount, as part of a changing climate, have the potential to impact future human activities in many areas. The influence of the change in anthropogenic aerosol concentration on precipitation is a significant uncertainty that needs to be better understood (e.g., Khain 2009). Improving the representation of precipitation in numerical models is a high priority, and it is a challenging task because many different processes can affect the onset, intensity, and duration of precipitation, including cloud and aerosol microphysical processes, dynamical processes, and thermodynamic state. The ASR program will use the new ARM Facility observational tools to characterize and quantify these processes so that they can be better represented in numerical models.

The atmospheric processes that govern the interaction of aerosol, clouds, and precipitation within the radiative, thermodynamic, and dynamical environment occur on a range of time and space scales. Thus, models with different characteristic scales (Figure 4) are used to study these processes. The processes

that initially determine cloud properties occur on scales that can be fully resolved only by fine-scale models. Fine-scale models are an important component used to help construct “parameterizations” (e.g., simplified representations of the fine-scale processes) that can be implemented in coarser-scale models. The evolution of physical processes, both in fine-scale and in larger-scale models, is complex and intertwined, and improving GCM predictive capability requires that both fine-scale and coarse-scale models be evaluated with suitable observational data sets. ASR observations are uniquely suited to help improve and validate models at smaller scales, and thus provide confidence that the parameterizations built from these fine-scale models are more accurate representations of reality.

The observational data sets required to evaluate and improve the understanding and modeling of atmospheric processes must capture the relevant processes that are at work in the climate system, as well as the different modes of variability. For example, some atmospheric processes may be more important, from a relative point-of-view, in different climatic regimes. Thus, it is important to collect data sets in these different regimes in order to sample the wide range of climatic conditions that must be represented in GCMs; this has been the motivation for the placement of the fixed ARM Facility sites in different climatic regimes (DOE 1991) and drives the selection of the past and future deployments of the airborne and mobile facilities (Figure 1). At each of these locations, it is important to make observations that capture the primary modes of variability, such as the diurnal, synoptic, and seasonal cycles. The ARM Facility stresses the deployment of state-of-the-art sensors that are able to collect data autonomously 24 hours per day, every day, as well as the development of the analysis techniques to derive the needed geophysical variables from these raw instrument observations. The ARM Facility’s continuous measurements, especially when supplemented by targeted laboratory studies and periodic intensive observational periods that include airborne in situ observations, provide the foundation for the process-study research that is needed to advance the capabilities and accuracy of models at a variety of scales up to and including GCMs (Fast et al. 2009).

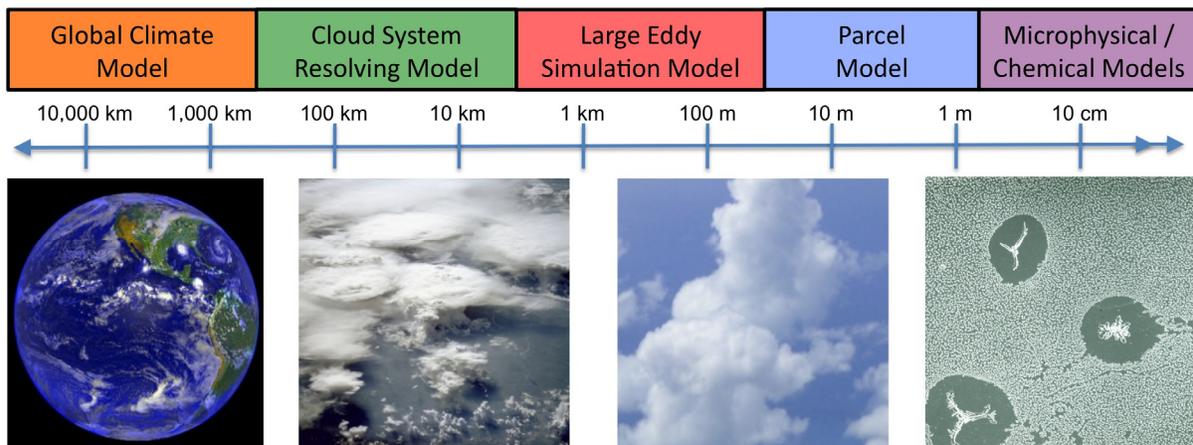


Figure 4. The various scales of numerical models that are used in climate science research by ASR investigators. Modified from S. Krueger.

1.3 Programmatic Strategy

Mission and Objectives

The goal of ASR, in partnership with the enhanced ARM Facility, is to quantify the interactions among aerosols, clouds, precipitation, radiation, dynamics, and thermodynamics to improve fundamental process-level understanding, with the ultimate goal to reduce the uncertainty in global and regional climate simulations and projections. To accomplish this goal, the ASR program and the ARM Facility will:

- Maintain and augment the collection of comprehensive and continuous long-term data sets that provide measurements of radiation, aerosols, clouds, precipitation, dynamics, and thermodynamics over a range of environmental conditions at several fixed and mobile sites situated in climatically diverse locations.
- Supplement the long-term data sets with laboratory studies and shorter-duration field campaigns, both ground-based and airborne, to target specific atmospheric processes under a diversity of locations and atmospheric conditions.
- Use these data, together with models, to understand and parameterize the processes that govern the atmospheric components and their interactions over all pertinent scales.
- Develop integrated, scale-bridging testbeds for model parameterizations that incorporate this process-level understanding of the life cycles of aerosols, clouds, and precipitation in numerical models.

A tight coupling of the enhanced ARM Facility and ASR program will allow the atmospheric system to be better observed and understood in a comprehensive, end-to-end fashion. The new ASR program is designed in recognition of the system as an aerosol-cloud-precipitation continuum operating within a microphysical and macrophysical environment characterized by radiation, dynamics (including meteorology), and thermodynamics. That continuum stretches seamlessly across scales from gases and primary particles emitted to the atmosphere, through evolving aerosol populations, to the clouds that form on aerosol particles, and the cloud systems that produce precipitation to complete the hydrologic cycle. The defining objective of the ASR program is a detailed, process-level understanding of this system that leads to improved simulations by climate models.

The aerosol-cloud-precipitation continuum, its role in the climate system, and the related processes that ASR aims to understand, are depicted in Figure 5. This figure is meant to illustrate many climate system properties and processes that pose difficulties to climate models and comprise the core focus of ASR research objectives; it is not meant to be comprehensive. Fundamental to the system are the life cycles of aerosols and clouds, and the interactions among aerosols, clouds, and precipitation. The term “life cycle” denotes the set of processes, indicated schematically in Figure 5, that govern the properties of atmospheric aerosols or clouds over time and that influence aerosol-cloud-precipitation interactions.

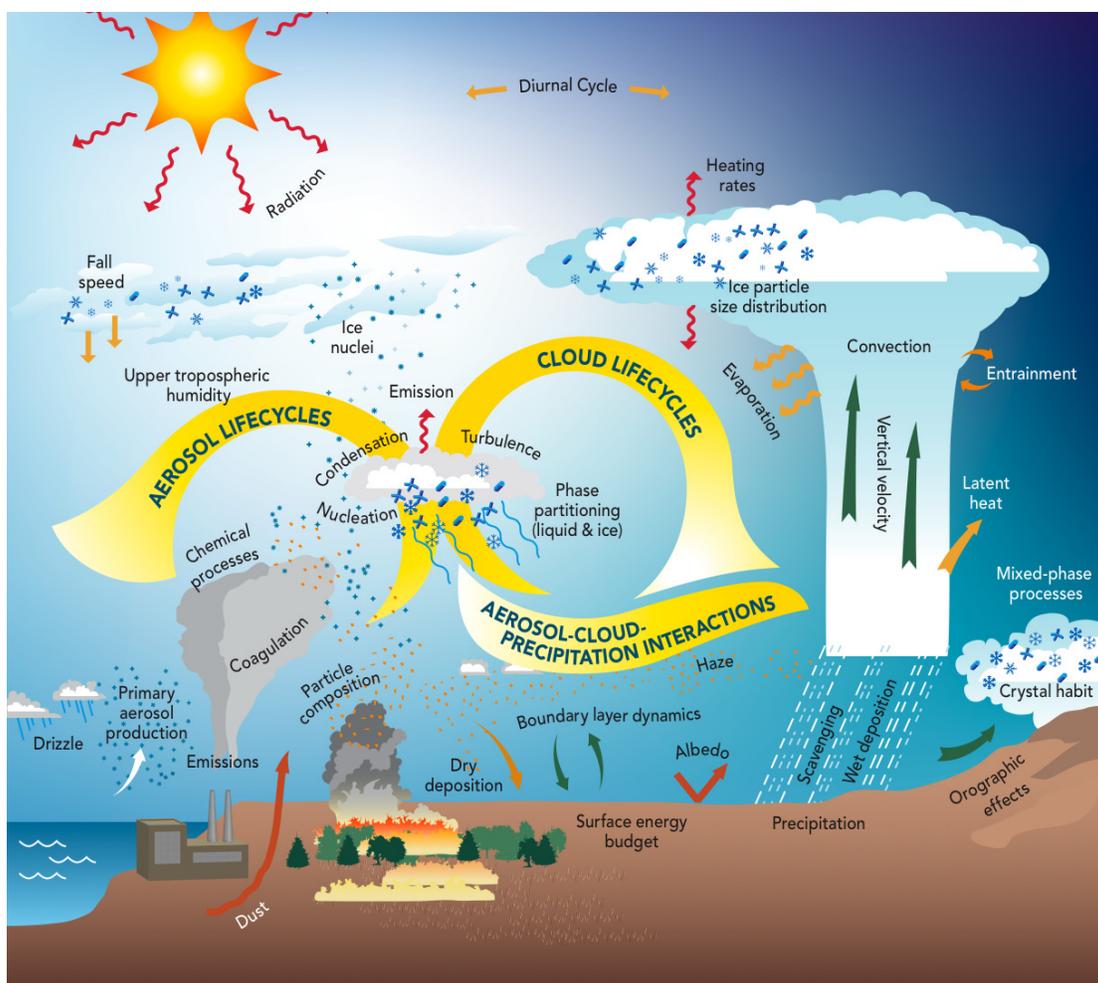


Figure 5. Atmospheric system depiction showing a subset of the atmospheric processes that must be understood and accurately represented in climate models. This figure schematically displays overarching themes (in all capital letters) and a representative, but not exhaustive, set of subsidiary processes that will be studied by the ASR program.

Aerosols are introduced into the atmosphere through numerous natural and anthropogenic pathways that fall into two general categories: direct emissions (e.g., soot, biomass burning smoke, and wind-blown dust) and new particle formation from anthropogenic and biogenic gas-phase precursor compounds (e.g. sulfur dioxide from power plants and naturally-emitted hydrocarbons from forested regions). Once present in the atmosphere, aerosol particles grow via reactive and non-reactive condensation of ambient gases and through coagulation, enhancing their optical properties and ability to serve as condensation sites for cloud droplet and ice particle formation. Although some aerosol particles exit the atmospheric system via sedimentation directly to the surface (dry deposition), most particles in the size range affecting atmospheric radiation and cloud microphysical properties undergo interaction with clouds and are ultimately removed from the atmosphere by precipitation.

The life cycle of a cloud begins when dynamic conditions generate a relative humidity in excess of the saturation vapor pressure required for the formation of cloud droplets or ice particles. Depending upon environmental conditions (e.g., temperature, dynamical state, and aerosol properties), cloud condensate forms as liquid droplets or ice crystals in a variety of shapes. The properties of the aerosols that serve as

seed particles for cloud formation play a central role in determining the cloud microphysical properties, specifically the number concentration, size, and phase. Microphysical processes, such as vapor deposition and particle collisions, which transfer water among the gas, liquid, and solid phases, lead to hydrometeor growth that can further influence cloud microphysical properties. Hydrometeor fall speed, which is dependent upon microphysical properties, is an important link in this cycle that controls cloud lifetime. The fate of clouds is governed largely by dynamical conditions, ranging from entrainment of drier air that leads to complete evaporation of small fair weather cumulus clouds to the self-perpetuating formation of storm cloud systems, which produce large hydrometeors that precipitate to the surface and extensive anvils that may dissipate over long periods of time. Throughout their life cycles, clouds exert a strong influence on the radiative fluxes at the surface, within the atmosphere, and at the top of the atmosphere.

At the confluence of these cycles, aerosol and cloud processes couple to give rise to the important but highly uncertain indirect effects that aerosols have on climate through their ability to modify cloud and precipitation properties. For example, a greater number of condensation nuclei leads to a higher concentration of smaller cloud droplets which, all other things being equal, results in a brighter cloud and one less likely to produce precipitation. This type of aerosol influence on cloud microphysical properties has profound impacts on radiative transfer, precipitation efficiency, and cloud lifetime. In a complementary fashion, clouds act to process aerosol particles; droplets can serve as sites for chemical reaction (e.g., oxidation of sulfur dioxide to sulfate) and as an important aerosol sink via particle removal through precipitation. Clouds are also the principal transporter of aerosols and aerosol precursor gases from the surface, where they are concentrated near local emissions sources, to elevations high in the troposphere, where winds transport them globally.

Diurnal, synoptic, and seasonal variability are manifested in many aerosol, cloud, and precipitation processes. These range from the daily heating and cooling of land surface, which influence cloud formation, to sunlight-driven photochemical reactions that are often a key step in aerosol formation. The latitudinal variation of these processes is also important, stretching from the steady diurnal cycle and minimal seasonal cycle in the tropics, to the continually varying diurnal cycle and strong seasonal cycle at high latitudes. A thorough understanding of how aerosol-cloud-precipitation processes respond and vary throughout these cycles is critical to resolving uncertainties in their impact on the climate system. ASR's use of the ARM Facility's long-term, automated observational capability will allow these important modes of variability to be investigated and understood.

Radiation and precipitation accumulation reveal the overall impact of the aerosol-cloud-precipitation continuum on the climate system. Both clouds and aerosols play key roles in atmospheric radiation through absorption, emission, and reflection of radiant energy in the shortwave (solar) and longwave (thermal infrared) spectral regions. Clouds and aerosol reflect nearly 80% of solar radiation that is reflected back to space. Atmospheric radiation absorbed by the surface drives surface temperatures and the freezing or melting of ice sheets, sea-ice, and glaciers. Radiation plays a role in defining atmospheric stability through differential heating, which impacts cloud formation and large-scale circulation. Global distributions of precipitation profoundly impact humanity and ecology by shaping the hydrologic and carbon cycles. Likewise, diabatic heating associated with precipitation formation is a major component of the global energy system (represented in Figure 4 by the latent heat component). Thus, with so many prominent roles in global climate, it is clear that even small uncertainties in modeling the aerosol-cloud-precipitation continuum can have a dramatic impact on simulated climate. In order to improve model skill in predicting the Earth's radiant energy and hydrologic cycles, it is therefore imperative to

understand and specify the controlling processes, which are closely tied to the vertically- and horizontally-resolved distribution of aerosols, clouds, and water vapor. Thus, accurately simulating the radiative field and precipitation in models is a necessary (albeit not sufficient) condition for demonstrating that processes associated with the cloud and aerosol life cycles, and the interaction between them, are properly represented in numerical models.

The ASR program will address the uncertainties in modeling the various processes in the aerosol-cloud-precipitation continuum. ASR's strategy towards characterizing this continuum in order to improve climate simulations is based upon a well-conceived suite of state-of-the-art instrumentation, measurement techniques, and experiments that are designed to observe fundamental aspects of the climate system at a variety of key locations. These observations target time scales ranging from rapid processes, through the diurnal evolution of the system, to seasonal and interannual variability of climatic conditions. Synergetic, multi-instrument methods are used to derive a comprehensive set of geophysical parameters that inform our process-level understanding, thereby providing the foundation for model development. Detailed process models are used to test and verify our physical understanding and, in turn, are essential for translating the results from local measurements to the much larger scales of climate models. Lastly, climate model simulations are evaluated against measurements and derived geophysical properties to confirm that the strategy has accomplished a reduction of model uncertainty, which is essential if the models are to accurately represent climate responses to putative future changes in atmospheric composition. Such a strategy is made possible by the timely blending of ARM and ASP resources, which originally focused on distinct portions of the atmospheric process continuum, and the addition of new ARM Facility instrumentation through ARRA.

2.0 Process Research

The climate research community is increasingly recognizing the tight coupling of clouds, aerosols, precipitation, radiation, thermodynamics, and dynamics as a single system that is interwoven with myriad linking processes. As a consequence, it is imperative to study these components of the climate system as a continuum, rather than as isolated elements. For this reason ASR will make the observations and perform the analyses necessary to develop, evaluate, and improve model parameterizations of this system. The ultimate objective is to leverage these activities towards increasing the accuracy of representations of aerosols, clouds, precipitation, and radiation in climate simulations. With the formation of the new ASR program, there is a remarkable convergence of long-term measurements, new instruments, and new approaches that will be harnessed to advance the observation and modeling of key atmospheric processes. Using these resources, scientific research conducted by the ASR program will target the overlap of (1) climate system processes and properties of the aerosol-cloud-precipitation continuum that are poorly represented in GCMs and (2) climate system processes and properties that are best observed using the ARM Facility ground, aircraft, and laboratory facilities, as well as those facilities of collaborating programs.

The primary research objectives for the new ASR program are presented in this section, grouped according to the themes of *aerosol life cycle*, *cloud life cycle*, and *aerosol-cloud-precipitation interactions*. In addition to fulfilling these objectives, research within the program will reach beyond these topics when required in order to include aspects of the natural environment that influence aerosol and cloud processes and couple them to the broader global climate system. To successfully understand and model the important processes comprising the aerosol-cloud-precipitation continuum, ASR will study and understand cross-cutting variables and processes, of which the most important are atmospheric thermodynamic profiles, surface turbulent heat fluxes, radiative transfer, and surface albedo, as well as how these variables and processes respond to diurnal, synoptic, and seasonal variability.

The ASR program's process-related research objectives:

- Determine the properties of, and interactions among, aerosols, clouds, precipitation, and radiation that are most critical to understand in order to improve their representation in climate models.
- Ascertain the roles of atmospheric dynamics, thermodynamic structure, radiation, surface properties, and chemical and microphysical processes in the life cycles of aerosols and clouds, and develop and evaluate models of these processes.
- Identify and quantify processes along the aerosol-cloud-precipitation continuum that affect the radiative fluxes at the surface and top of the atmosphere and the radiative and latent heating rate profiles, and improve the ability to accurately model these processes.

2.1 Aerosol Life Cycle

Aerosol particles exert important influences on climate and climate change by scattering and absorbing solar radiation and by influencing the properties of clouds. The aerosol life cycle determines the spatial and temporal distribution of atmospheric particles and their chemical, microphysical, and optical properties. The objective of aerosol life cycle research in ASR is to improve understanding of the roles of aerosols in the climate system and specifically to decrease uncertainty in radiative forcing by aerosols. The processes associated with the life cycle of aerosols must be understood and represented in climate models, first, to represent the unperturbed climate with sufficient fidelity to obtain an accurate model-based determination of the sensitivity of climate to radiative perturbations locally and globally, and second, to determine the radiative forcing of aerosols, i.e., the changes in radiative budget at Earth's surface and at the top of the atmosphere due to perturbations in the amount and nature of aerosols. Current aerosol models need improvement in several areas: emissions; mechanisms, and rates of new particle formation and their dependence on controlling variables, growth, and aging; distinguishing natural and anthropogenic influences; and computational efficiency of model representation. In addition, the models need to be systematically evaluated to determine the cause of differences between simulated and observed aerosol properties. The research that will be conducted by ASR to achieve these objectives is usefully distinguished into four topical areas or elements: new particle formation, aerosol growth and aging, the direct radiative impacts of aerosol, and separating the natural vs. anthropogenic aerosol influences on aerosol properties.

2.1.1 New Particle Formation

Besides primary emissions of primary aerosol particles, another important source of atmospheric aerosols is the *chemical conversion of gas-phase atmospheric precursors* to new particles through nucleation and growth. Freshly nucleated particles are approximately a nanometer in size and thus are too small to directly influence climate. But these particles can quickly grow, even over the course of a single day, to reach the size where they can serve as cloud condensation nuclei (CCN) or appreciably scatter light. Understanding the molecular processes leading to the nucleation and growth of new particles is a current objective of field observations, laboratory measurements, and theoretical study. The field observations show that boundary layer nucleation rates can be orders of magnitude higher than were calculated by early models and suggest that other atmospheric trace species, besides water and sulfuric acid, which were already included in the early models, contribute to these high rates. Candidate tropospheric precursors include ammonia, amines, and organic acids, which have been shown in laboratory and theoretical studies to act in concert with sulfuric acid-water through ternary nucleation mechanisms to enhance nucleation rates. To improve understanding of new particle formation and its dependence on precursor gases and existing aerosol, field studies combining measurements of the size and composition of molecular clusters and small particles with measurements of concentrations of key precursor gases will be useful, as well as laboratory studies that systematically examine dependence of new particle formation rates on specific trace gases.

A strong coupling between nucleation and growth rates of nucleated particles further contributes to the climate importance of new particle formation. *Growth rates of nucleated particles* are commonly five to twenty times higher than can be explained by the condensation of sulfuric acid vapor alone, and these high rates enhance the probability that nucleated particles grow to sizes that are large enough to serve as CCN. Recent work has shown that the fast growth rates are due largely to the uptake of oxygen- and

nitrogen-containing organic compounds. Future work on new particle formation will focus on developing models for the mechanisms by which organic compounds enhance nucleation and growth. Additional importance derives from the fact that reliable growth models are also needed to solve the outstanding problem of explaining observed amounts of secondary organic aerosol formation. Laboratory and field measurements are needed to develop empirical models of new particle formation and growth for immediate use in models and to test new theoretical models that explain the measured dependence of new particle formation rates on atmospheric gas-phase precursor and background aerosol concentrations.

2.1.2 Aerosol Aging and Mixing State

Aging of aerosols consists of modification of the composition, size, and surface properties of aerosol particles in the atmosphere by coagulation, condensation, and surface reactions. The concept of aging is also sometimes used to refer to the mixing of aerosols from different sources to form more complex external mixtures. These processes are important as they affect the optical and cloud nucleating properties of the aerosol. Consequently these processes must be understood and represented in models. ASR will examine these processes in field studies and in laboratory experiments that are informed by the field measurements to develop process models that can represent these phenomena in large-scale aerosol chemical transport models and ultimately in climate models.

Measurements downwind of urban sources of aerosol particles and precursor gases have shown that the mass concentration of *secondary organic aerosol* (SOA) can be several-fold greater than can be explained on the basis of current model calculations using observed precursor concentrations. ASR will continue conducting laboratory experiments on both gas-phase and aqueous-phase SOA formation to characterize the particle formation and the organic gases that react to form new organic aerosol material on aerosol seeds. The dependence of SOA formation on nitrogen oxides and other factors will be examined. ASR will use these experiments to guide the development of comprehensive chemical mechanisms, and then use improved understanding from the comprehensive mechanisms to guide the development of parameterizations that are simple enough to be applied to aerosol life cycle models. To test the models under real world conditions, a series of field studies is needed to improve understanding of the interactions between natural and anthropogenic SOA formation mechanisms.

Secondary aerosol formation can profoundly change the *optical and microphysical properties of particles* when the composition of the primary particle is very different from that of the secondary material. Of particular importance is the aging of black carbon particles due to both condensation of secondary material and coagulation with other particles. ASR will continue to conduct laboratory experiments and field measurements to characterize the influence of aging on the optical and microphysical properties of black carbon particles. Data from these studies can be used to test models of how the aging process affects the optical and microphysical properties of black carbon. SOA can also absorb both shortwave and longwave radiation and characterization of its optical properties and relative contributions from natural and anthropogenic sources are also currently areas of active ASR effort.

Aged aerosol often includes a composite of mixtures of particles of different compositions along with species condensed through condensation. These general *mixing states* poses challenges for a simple representation in aerosol life cycle models, as the mixing state and how aerosol ages under different conditions impact aerosol direct forcing and CCN concentration. Because it is not practical to track the size, shape and composition of every single particle in a global model, approximations must be

introduced. One approach to this challenge uses single particle measurements of size, shape and composition during laboratory experiments and field studies of aging to test both particle-resolved and simplified representations of the aerosol mixing state. The goal is to develop representations that are more flexible than current treatments, which assume all particles of the same size have the same composition, without resorting to tracking every particle. The figures of merit will be the ability of the representation to reproduce measurements of CCN concentration and aerosol optical properties. As climate models require great economy of aerosol representation it is advantageous to seek the minimum number of tracers required to meet this criterion.

2.1.3 Aerosol Direct Radiative Forcing

Scattering and absorption of solar radiation by aerosols (so-called aerosol direct radiative effects) modify the amount of incoming solar radiation taken up by Earth, and modify the vertical distribution of that absorption and the resultant heating profile of the atmosphere. Uncertainty in the scattering and absorption of radiation by aerosols in cloud-free air contributes substantially to the uncertainty associated with aerosol radiative forcing of climate change over the industrial period. This uncertainty is largely due to high frequency variability in aerosol optical properties in space and time and the lack of methods available to measure them at high resolution globally. Accurate representation of aerosol direct radiative forcing in climate models requires that aerosol optical properties be properly determined from the size distributed aerosol composition, a relationship that is presently not well understood.

Aerosol optical properties depend strongly on particle size, composition, and morphology. Furthermore, many aerosol constituents are hygroscopic, meaning that they readily uptake water; thus with increasing relative humidity the radii of the aerosol particles increase, resulting in a corresponding change in other aerosol properties, such as index of refraction that affects the scattering and absorption coefficients of the particle. Thus, direct aerosol forcing is strongly dependent on relative humidity. As the variability of aerosol properties with relative humidity is likely greater than any other quantity (such as temperature), quantifying and parameterizing the *dependence of aerosol properties on relative humidity* is the key to accurate representation of this process in models.

To assess and improve understanding and representation of aerosol effects on the Earth's energy balance, ASR will use both local and column *radiative closure* for long periods (months) at multiple locations. Local closure will compare the measured aerosol optical properties (absorption, extinction and asymmetry parameter) with model calculations given the measured relative humidity and the size-resolved shape and composition of the aerosol. Such a study will require advances in the ability to measure aerosol absorption, shape and composition for all particle sizes important for radiative closure. Column closure will use 3D surface-based remote sensing and in situ measurements from aircraft to characterize the clouds and aerosol above surface measurement sites, and then compare the measured radiative fluxes at the surface and top of atmosphere with those calculated by radiative transfer models.

Aerosols containing black (elemental) carbon and organic carbon that absorbs in the visible spectral region have a potentially large impact on climate by reducing the net solar radiative flux at the Earth's surface and by warming the air in their vicinity, where the latter can lead to cloud evaporation (referred to as the aerosol "semi-direct effect"). To date, quantifying these effects has proved difficult. Recent laboratory studies have examined the aging properties of black carbon aerosols whereby particles become encapsulated by coatings that can induce compaction, which decreases light scattering and absorption,

and lensing, which is an effect in which more light is focused by the coating thereby increasing the light absorption. Understanding this interplay between morphology and optical properties is necessary to quantify the contribution of *light absorbing carbon* to direct and semi-direct radiative forcing. Aging also affects the ability of black carbon aerosols to serve as cloud condensation nuclei through modification of their ability to become wetted and condense water. Examination of these phenomena will continue to be a focus of research in ASR laboratory experiments and field measurements.

2.1.4 Natural vs. Anthropogenic Influences on Aerosol Properties

The need to understand atmospheric aerosols and their influence on climate and climate change is driven by the requirements to (1) represent these influences in models of the present climate, thereby accurately simulating aerosol effects on clouds and radiation in the present climate; and (2) to represent the influences of changes in the amount and properties of aerosols (over the industrial period, or for prospective future emissions) on atmospheric radiation and cloud properties, in order to permit examination of past and prospective future climate change. For these reasons it is necessary to measure and model natural as well as anthropogenically influenced aerosols and to distinguish the climate influences of each. Measurements that *distinguish natural and anthropogenic influences* on aerosol properties are needed to determine the anthropogenic perturbation to radiative forcing. This, in turn, helps to unravel the extent to which anthropogenic aerosol perturbations, which tend toward cooling, mask the effects of greenhouse gas warming. Generally, both anthropogenic and natural processes contribute to the atmosphere aerosol, influencing the new particle formation, the role of anthropogenically enhanced levels of atmospheric oxidants in SOA formation from natural and more volatile organic emissions, and the oxidation reactions on aerosol surfaces and within cloud droplets that result in increases in aerosol mass and hygroscopicity. ASR research will examine the contributions of anthropogenic versus biogenic organic aerosol precursors and determine the chemical mechanisms responsible for the conversion of these compounds to increase aerosol number and mass and for new particle formation and aerosol dynamics. While most current climate models attempt to separate natural and anthropogenic aerosol influences, they tend to miss some important interactions. A promising approach is to distinguish natural and anthropogenic contributions to aerosol loading through tracer measurements (e.g., carbon isotopes) for model testing.

2.2 Cloud Life Cycle

Approximately 20% of the solar radiation incident on Earth is reflected back to space by clouds, thereby preventing absorption that would otherwise warm the surface. It is therefore not surprising that small uncertainties in the representation of cloud properties in GCM simulations (e.g., cloud cover, precipitation, structure, lifetime, or reflectivity) have the potential to induce large uncertainties in model computed surface temperatures and other important climate properties. Many of the uncertainties in GCMs stem from poor representation of cloud processes that operate at fine scales. Important examples are lateral and cloud-top entrainment and drizzle formation, which influence the reflectivity and persistence especially of marine stratocumulus clouds, a climatologically important cloud type, the representation of which has been identified as a major source of variation in sensitivity of GCMs (e.g., Bony and Dufresne 2005). ASR will study these and other cloud life cycle processes over the full range of relevant space and time scales. Specific cloud life cycle research areas are categorized into three broad areas: *dynamics*, the atmospheric motions that generally dominate cloud life cycles; *microphysics*, the properties of cloud droplets/ice particles and rain and snow hydrometeors and the processes that

determine these properties and their interactions; and *radiation*, the impacts of cloud amount and properties on absorption, emission, and transport of shortwave and longwave radiation in the atmosphere, at the surface, and at the top of the atmosphere. The important problems and processes that ASR will target related to each of these broad categories are outlined below.

2.2.1 Dynamics

Vertical air motions play a central role in cloud life cycles. Whereas emissions and nucleation lead to new aerosol particles, it is primarily atmospheric dynamical conditions in a sufficiently humid environment that lead to new cloud particles. To simulate cloud life cycles, models must adequately represent the strength and depth of updrafts and downdrafts. However, vertical velocity of cloudy air is difficult to measure, and modelers desire long-term measurements of vertical air velocity statistics to develop and evaluate cloud parameterizations. In response, ASR will pursue multiple research fronts towards better understanding vertical air velocity, its relation to buoyancy forces, and the associated dynamical-microphysical interactions. These concepts will be studied in all types of clouds including, but not limited to, shallow marine cumuli, Arctic stratocumulus, frontal clouds, orographically forced clouds, deep convective storm systems, and upper tropospheric cirrus. Two specific cases are discussed here as an example. The first addresses shallow circulations associated with small cumulus and stratocumulus clouds, which are typically driven by changes in atmospheric buoyancy as a result of surface turbulent energy fluxes and/or radiation. Important parameters in such clouds include vertical air velocity variability, the structure of turbulent motions, the skewness of the vertical air velocity distribution, and atmospheric stability profiles. ASR will use a multi-instrument and -platform approach to study the co-variability of these dynamical parameters with the cloud microphysics in order to reveal the important linkages between boundary layer dynamics, radiation, cloud formation, and cloud composition, all of which are crucial to understanding the life cycle of these clouds. A second example of ASR research is focused at the other extreme of size, where organized deep convection associated with storms is characterized by large-scale structural organization that can be categorized into convective and stratiform fractions. In these cases, ASR will strive to understand how the updraft and downdraft properties, spatial extent, depth, and structural organization impact the amount of precipitation, the vertical distribution of condensate, and the microphysical composition of resulting cirrus.

Entrainment of environmental air into clouds is a key process that is poorly represented in GCMs. In convective clouds, the rate of entrainment of ambient air into convective updrafts directly affects the buoyancy of those updrafts and thus can strongly influence the vertical velocity and the depth of convective updrafts, thus controlling cloud vertical extent. Current entrainment parameterizations are empirical, as entrainment is a process that is difficult to directly observe. Entrainment rates are typically inferred using detailed measurements of environmental conditions and theoretical or model calculations to deduce the mixing that is required to explain them. Turbulence probes flown on research aircraft have provided some validation of underlying entrainment mechanisms; however, due to the complexity of the entrainment process and the difficulty in locating entrainment regions, samples are often small and/or inconclusive. Additional process-level observations, such as cloud-scale vertical velocity, are sorely needed to better constrain and improve entrainment parameterizations. Because the impact of entrainment on cloud buoyancy depends strongly on the humidity of the air entrained into a cloud, accurate retrievals of water vapor in the near-cloud environment are needed. In particular, ASR will focus on understanding entrainment processes in shallow and layered clouds, which have a relatively thin entrainment layer that can have large impacts on the cloud persistence and microphysical composition. In

addition, entrainment and detrainment processes in convective cloud systems will be studied to better understand convective strength, precipitation development, and the vertical transport and distribution of atmospheric moisture, particularly in the upper troposphere.

Convective initiation marks the beginning stage of the cloud life cycle where many specific processes converge. While vertical air motion and entrainment play fundamental roles, many other 3D phenomena affect development of clouds and storms, including lifting over topographic features, boundary layer and tropospheric thermodynamic structure, surface turbulent flux inhomogeneities (e.g., plumes of warm air rising from sunlit surfaces), and gust fronts and density currents associated with cloudy downdrafts. Due to this variety of structurally complex factors, incorporating convection initiation mechanisms into convective parameterization for weather forecast, regional, and global climate modeling is an extremely difficult, yet profoundly important, task. GCM-simulated precipitation frequency, intensity, and temporal variability are quite different from those observed and these can be traced to inadequate parameterization of convection triggering. From an observational perspective alone, it is very difficult to unravel the multiple effects on cloud evolution. ASR will strive to develop a unified understanding of the convergence of processes associated with triggering convection through the focused coordination of measurement and modeling efforts.

2.2.2 Microphysics

To truly understand cloud life cycles and the multifaceted way in which clouds interact with the climate system, it is imperative to characterize the *cloud particle size distributions*, which describe the number of particles of each type and size. Once dynamics are accounted for, it is the particle size distributions, and their interaction with radiative processes, that determine a cloud's evolution and fate. Accurate knowledge of the hydrometeor number, size, surface area, volume or mass, dispersion, skewness, and phase are required in order to understand basic cloud processes such as the microphysical evolution through competition for available water vapor, formation of precipitation-sized particles, sedimentation, and collisions among cloud particles. Two specific cases are outlined here as examples. First, the relative concentration of small ice crystals in the ice particle size distribution is unknown due to various observational limitations. However, it is extremely important to characterize the small crystal distribution as it can exert a potentially significant control on the radiative effects of ice clouds. Second, within mixed-phase clouds both liquid and ice size distributions exist within the same cloud system, interacting and coevolving through myriad, complex mechanisms. A clear accounting of both solid and liquid hydrometeors in these clouds is needed to determine their radiative effects, precipitation efficiency, and longevity. It is a primary goal of the ASR program to characterize the particle size distributions for all types of clouds in as broad a set of conditions as possible, including precipitating conditions, in order to adequately represent these size distributions and their evolution in cloud process models.

A full characterization of the ice particle size distribution is complicated by the variety of *ice crystal habits* present in the atmosphere. Whereas droplets are spherical and deviations of rain drops from sphericity are relatively well known, ice particles exist in a dizzying array of habits that influence fall speed, growth, and evaporation rates, as well as the efficiency of collision and coalescence. Modelers struggle to efficiently represent the evolution of ice habit. However, the range of size-distributed habit complexity that exists in the atmosphere and the most efficient means of adequately representing it in climate models remain unknown. The ASR program will accelerate progress towards understanding the

role of ice crystal habit by better characterizing ice habit distributions (e.g., habit identification, crystal population diversity as a function of size, and projected area) and elucidating the impact that habit has on microphysical and radiative processes of cloud ice.

Ice crystal fall speed has long been known as a remarkably powerful control on climate model simulations, and one that remains very poorly constrained by measurements. The fall speeds of liquid-phase particles are relatively well understood over the full range of sizes in the atmosphere; however, ice fall speeds are greatly complicated by the myriad habits and resultant densities that occur. Advances in understanding crystal fall speed will be achieved through a specific focus on making detailed collocated measurements of ice crystal habits, particle size distributions, radar reflectivity, and Doppler velocity spectra.

As hydrometeors grow via vapor deposition, collision, and coalescence processes, they eventually reach large sizes that fall rapidly and are responsible for the primary flux of condensed water from the atmosphere to the surface. Thus, an understanding of *precipitation formation* processes is vital for determining the role that the atmospheric system plays in the hydrologic cycle. Precipitation formation processes remain poorly understood in both warm (liquid-phase) and cold (ice-containing) clouds. In cold clouds, aerosol-induced heterogeneous droplet freezing and ice crystal nucleation are poorly characterized. In warm clouds, several precipitation formation theories have been proposed, placing emphasis on giant aerosol particles, turbulence-induced coagulation, or turbulence-induced fluctuations in saturation and droplet growth. These hypotheses are extremely difficult to examine owing to the multitude of processes that can contribute to precipitation formation. The ASR program will better characterize precipitation properties and rates, along with their relation to dynamic and thermodynamic states, by examining the role of different hydrometeor growth mechanisms and better constraining parameters such as vertical velocity. In addition, ASR will characterize clouds and precipitation together to understand the linkages between the microphysical properties of clouds and the precipitation they produce.

2.2.3 Radiation

The amount of solar radiation that reaches the Earth's surface is strongly influenced by *cloud optical depth*. ARM Facility measurements under clear-sky conditions indicate that the shortwave and longwave spectral and total surface fluxes can be calculated to 1-2% accuracy if the spectral surface albedo and the vertical profiles of water vapor, ozone, temperature, and aerosol optical properties are known. Climate models require calculations of radiation to similar accuracy under all conditions, not just clear sky; however, this degree of accuracy has not yet been realized. Most cloud radiation research has been limited to the simplest case of overcast cloud conditions where 1D (i.e., plane-parallel) radiative transfer assumptions are met. These conditions occur, for instance, less than half of the time at the ARM Facility's mid-latitude Oklahoma site. A more general representation of cloud radiative effects requires knowledge of the 3D cloud extinction structure, whose simplest manifestation of is cloud optical depth, the vertical integral of extinction. Through the combined retrieval of cloud particle size distribution properties and 3D cloud mass concentration, ASR will work to directly characterize 3D extinction structure and thereby minimize the overall uncertainty in cloudy-sky radiative transfer calculations.

Recent studies have highlighted that, in many areas of the globe including at some of the ARM Facility fixed sites, the surface solar radiative flux has changed over the last several decades. The mechanisms

behind this “*dimming and brightening*” phenomenon are not understood, but various hypotheses include a change in the aerosol size distribution, changes in cloud properties, or changes in upper tropospheric humidity. Dimming and brightening should be intimately related to the climatological connections between radiation and cloud and aerosol processes. The ASR program will leverage the enhanced ARM Facility suite of instruments to better understand the long-term changes in surface radiation and how clouds and aerosols impact the radiative field.

An important cloud-radiation focus is the *spectral dependence of ice absorption* in both cirrus and mixed-phase clouds, and the need to validate and improve single scattering property models across the entire spectrum from the ultraviolet to the far-infrared. The far-infrared (15–100 μm) and near-infrared (1–4 μm) are relatively underexplored due to lack of instrumentation that are able to make spectrally resolved observations in these bands, yet approximately 40% of the total outgoing longwave flux is from the far-infrared portion of the spectrum and 35% of the total shortwave flux at the surface is from the near-infrared. New instruments have recently been developed to make spectrally resolved observations in these bands in order to evaluate and improve both gaseous absorption models and cloud radiative properties in these spectral bands. To increase the accuracy of radiative transfer models in the near-infrared, the spectral surface albedo, precipitable water vapor, and aerosol size distribution (especially for super-micron particles) all need to be better characterized. ASR will strive to better constrain radiative transfer in these underexplored bands leading to improved radiation parameterizations for GCMs, as was done with earlier radiative transfer advancements made by the ARM Program.

The quantification of the *radiative heating rate profile* will continue to be an emphasis for ASR. Through their radiative interactions with the atmosphere, clouds affect atmospheric stability, which in turn feeds back to the cloud dynamical-microphysical interactions that are responsible for cloud formation. The cloud life cycle cannot be understood without also capturing these important feedbacks. Because climate models typically struggle to capture these feedbacks, they often poorly represent the cloud fields in general. Since radiative heating rate profiles are intimately tied to the microphysical properties of clouds and aerosols in the atmosphere, heating rate calculations also provide a means to validate cloud and aerosol property retrievals via radiative flux closure studies. Additionally, the ARM Facility’s new ability to make 3D cloud observations will allow for a more accurate computation of the domain-averaged radiative heating rate profile which can be used to characterize the error associated with using 1D radiative transfer approximations in lieu of a full 3D calculation in various cloud fields. The ASR program will develop a better understanding of 3D radiative transfer in both stratiform and cumuliform cloud fields as a function of domain size.

2.3 Aerosol-Cloud-Precipitation Interactions

Aerosols and clouds are inextricably coupled throughout their life cycles in processes that dictate cloud formation and development, spatial coverage, persistence, and precipitation efficiency. The radiative forcing associated with aerosol-cloud-precipitation interactions is referred to as the *aerosol indirect effect*. Since pre-industrial times, globally averaged, net, top-of-atmosphere mean radiative forcing by aerosols is estimated to be about 75% of that due to the increase in carbon dioxide, but of opposite sign. About 60% of this negative forcing is via the cloud-albedo effect, one of the specific aerosol indirect effects; however, the radiative forcing of the cloud-albedo effect remains highly uncertain. Several of the climate models participating in the IPCC 4AR did not include anthropogenic aerosol-cloud interactions owing to lack of process understanding and of reliable approaches to model representation. Of the representations

that were included, different parameterization approaches (physical and empirical) resulted in large differences in their ultimate radiative impacts. To better represent aerosol-cloud-precipitation interactions in climate models, ASR recognizes the critical need for improved understanding in several areas: (1) the impact of aerosols on cloud particle formation processes that affect the concentration and size-distribution of cloud particles, (2) radiative impacts determined by the microphysical and macrophysical structure of clouds influenced by aerosol, and (3) precipitation efficiency dictated by a myriad of related processes including cloud depth, collision/coalescence, and entrainment. Cloud processing of aerosol also plays an important role in aerosol chemical and microphysical properties through aqueous-phase chemistry, aerosol removal and vertical redistribution mediated by precipitation and vertical motions, and aerosol global characteristics and geographical distribution.

2.3.1 Cloud Particle Formation

A key requirement for simulating aerosol-cloud interactions is the ability to calculate *cloud condensation nuclei* and *ice nuclei* (CCN and IN, respectively) concentrations as a function of supersaturation from the chemical and microphysical properties of the aerosol. There is a need to quantify aerosol influences on cloud drop and ice particle concentrations, size distribution, ice crystal habit, precipitation development, and the dependence of these influences on the anthropogenic and natural aerosol components. Understanding of cloud particle formation processes coupled with known CCN and IN concentrations allows for direct simulation of cloud particle concentrations, one of the main determinants of cloud albedo.

Several key processes concerning *droplet formation* in warm clouds remain poorly understood. The influence of subgrid-scale convection and entrainment on supersaturation and droplet nucleation needs to be characterized so that aerosol effects on shallow cumulus clouds can be treated in climate models. Entrainment effects on droplet formation are represented in models, but have not been tested with long-term observations. The effects of aerosols on droplet dispersion need to be understood well enough to be expressed in a physically-based, rather than the current empirically-based, manner. Representing aerosol effects on precipitation and cirrus anvil detrainment from deep cumulus clouds requires understanding of the environmental factors that affect the sensitivity of deep convective clouds to aerosols. That understanding can lead to better treatments of aerosol effects on the microphysics, precipitation, and detrainment from parameterized cumulus clouds.

Representation of aerosol effects on *ice particle formation* will require improved understanding of both the aerosol properties that determine ice nuclei concentration as a function of saturation, and the processes that control the supersaturation in updrafts. ASR will use laboratory experiments to improve understanding and to develop parameterizations of ice nucleation, as well as field measurements of ice nuclei concentration and the size and composition of ice crystal residuals to evaluate the nucleation parameterizations. To improve understanding of supersaturation, cloud-resolving models with the ice nucleation, parameterizations will be used to determine the dynamical and microphysical processes that control supersaturation in cirrus and mixed-phase clouds. This improved understanding can in turn be used to develop subgrid-scale treatments of supersaturation, evaluated using field measurements from an array of new airborne aerosol, cloud, and atmospheric state probes.

2.3.2 Cloud Processing of Aerosol

Much of the atmospheric processing that governs the transformation of precursor gases into aerosol particles occurs in the aqueous phase of clouds, with resulting changes not just in the amount of aerosol but also in the size distributed composition. The *processing of aerosol by clouds*, including vertical transport, aqueous-phase chemistry, droplet collision/coalescences, and wet deposition can affect the distribution, composition, and size distribution as well as the mass concentration of the aerosol. The principal process by which accumulation-mode aerosol particles are removed from the atmosphere involves uptake of these particles into cloud drops which subsequently precipitate; thus precipitation development both depends on aerosol properties (through the influence of aerosols on cloud drop number concentration and size) and influences the amount and nature of aerosol particles that are deposited on the surface or returned to clear air upon cloud droplet evaporation.

Improved measurements of organic and inorganic components present in precipitation can aid in evaluation of aerosol lifetimes and *removal processes*. Indeed, the measurement of soluble organic materials in precipitation and their isotopic and molecular signatures may be a very useful means of determining aerosol sources as well as preferential removal of specific components in aerosols. Characterization of aerosol derived components in precipitation can shed light on the importance of hygroscopic aerosol species and soluble gases on cloud processing of aerosol, as well as providing information concerning the potential impact of light absorbing aerosols on clouds, as well as the geographical distribution of aerosol in cloud-free conditions.

2.3.3 Precipitation

Aerosol induced changes in the number and size distribution of cloud droplets or ice particles influence the *development of precipitation*. The sum of these and cloud dynamical processes dictates not only the amount of precipitation reaching the ground, with implications for water resources, but also the persistence and structure of the cloud, with implications for cloud radiative forcing. For example, the full interactions between clouds and aerosols in boundary layer stratocumulus are thought to cause the rapid transition of the cloud-aerosol system from relatively polluted closed-cellular convection with weak precipitation to clean open-cellular convection with more intense precipitation. This transition has profound implications for precipitation, as well as cloud field structure and cloud radiative forcing, and is a topic of ASR research.

A commonly accepted theory of aerosol-cloud-precipitation interactions in warm clouds is that as aerosol concentrations increase, cloud droplets become smaller, thus suppressing precipitation. A secondary effect is that clouds persist and may increase in spatial coverage. However, modeling studies show that contrary to expectation, an increase in aerosol concentration from very clean to very polluted does not increase cloud persistence in shallow boundary layer clouds, even though precipitation is suppressed. Processes such as evaporation-entrainment feedbacks tend to dilute polluted clouds more than clean clouds, creating competing effects of precipitation suppression and enhanced evaporation. Precipitation in cold clouds is usually initiated by ice formation. Additional IN can enhance precipitation from cold clouds by nucleating ice crystals, which grow by vapor deposition and collection of cloud droplets. The net effect on cloud albedo depends on the relative increase in cloud-activated CCN and IN. Accurate model representation of precipitation-related processes impacts cloud albedo through changes in

microphysics as well as cloud coverage and persistence. ASR scientists will investigate these *processes that contribute to cloud persistence* to ensure that the basic physical mechanisms are properly captured within models.

To advance understanding of precipitation processes in warm clouds, *turbulence and drizzle* measurements will need to be connected to the aerosol measurements to facilitate studies of the effects of CCN on cloud and drizzle properties, the effects of the wet removal of aerosol and/or evaporation of drizzle in modifying local aerosol, and defining cloud updraft statistics near cloud base where CCN are activated. In cold clouds, mass concentration and fall speed are basic parameters for ice and snow precipitation. Models also need to represent the *size distribution and crystal habit* properly, including factors such as cross-sectional area, density, and maximum diameter. In the Arctic, for example, habit and fall speed appear to be extremely important to the lifetime of the cloud and the evolution of the thermodynamic field. Improving model representation of ice cloud is essential for understanding precipitation and is currently a major focus area for model development at a range of scales.

2.3.4 Radiative Impacts

Anthropogenic increases in cloud-activated CCN modify cloud microphysical properties by decreasing the average cloud droplet size thus increasing the number of droplets sharing the same amount of cloud liquid water. This process is known as the *cloud albedo effect*. A reduction in the cloud droplet size can impact the atmospheric energy balance because the smaller droplets reflect more incoming solar radiation to space than clouds unaffected by aerosol and proportionally reduce the amount of radiation reaching the surface. This process is not limited in its radiative impact to just shortwave radiation, but has also been shown to impact infrared radiation especially in clouds with low amounts of liquid water path such as in the Arctic. Additional research is needed to more fully quantify both the shortwave and longwave albedo effect under a wider range of environmental conditions.

Quantifying the cloud albedo effect requires a statistical measurement approach in order to isolate it from the range of cloud dynamical mechanisms that may change the amount of cloud liquid water and hence the solar reflectivity. Measurements from ARM Facility surface-based remote sensors have been used to pioneer observing strategies used to examine and quantify specific *links between cloud liquid water, cloud droplet size, cloud dynamics, and cloud reflectance*. The program has also advanced understanding of aerosol indirect effects through the use of new, specialized instrumentation that enables the CCN feeding cloud updrafts to be accurately counted and the response of the cloud structure to be simultaneously gauged. Results derived from ARM Facility observations have shown that measurements of aerosol indirect effects are sensitive to the observing methodology, which is motivating ASR in the development of new approaches to address these sensitivities.

In addition to their influence on cloud microphysical properties, aerosols have a strong influence on *cloud formation and development* and thus dictate to some extent cloud distributions and cloud macrophysical properties. It has become increasingly evident that the size and distribution of clouds is an important factor influencing cloud albedo. In warm boundary layer cloud systems, small clouds have been shown to have the greatest contribution to cloud number, and large contributions to cloud area (fraction) and reflectance. Aerosol- and dynamical-related processes that influence cloud field structure are not well understood at this time and observational approaches have not been fully developed for the current suite of sensors. Small clouds are difficult to detect and quantify because they often fall below the resolution

of most space-borne observational sensors. ARM Facility observations at a range of scales are being used to improve understanding of cloud field structure and to use this information in 3D models to further progress in radiative transfer of broken cloud fields.

The light-absorbing properties of aerosols can have a strong influence on cloud dynamics though heating, referred to as the *semi-direct effect*. Typically, the presence of absorbing aerosol is thought to induce atmospheric stability, suppress vertical motion, and decrease cloud formation. However, the localized heating caused by absorbing aerosol may work to either stabilize or destabilize the boundary layer depending on interactions with surface properties and processes, dictating whether or not reductions in overall cloudiness occur.

Aerosols affect the Earth's energy budget primarily by affecting the global albedo. A continuously varying range of albedo values, with end-members of clear and cloudy, results from variable and currently indeterminate combinations of hydrated aerosol from sub-visual or tenuous cloud to optically thick cloud. Nevertheless, many research methods are premised on a dichotomy of clear and cloudy states, dividing the aerosol forcing problem into direct and indirect components respectively. Two consequences arise. First, observational approaches that rely on discrimination of clear versus cloudy data points are inconsistent and sensitive to variation in concentrations of unactivated aerosol particles. Second, aerosol radiative forcing is inaccurately calculated as the average of clear and cloudy conditions, which may lead to substantial errors resulting from neglect of the intermediate states of albedo. For example, an increase in direct radiative forcing over oceans in partly cloudy environments caused by enhanced scattering of hydrated aerosols near clouds, so-called "halos," may contribute substantially to average albedo but is ignored as a result of being classified as either clear or cloudy. Treating the direct and indirect components of aerosol radiative forcing holistically, and not as disjoint phenomena, will improve understanding and representation of the clear-to-cloudy range in global albedo.

3.0 Measurement and Process Modeling Research

Observations are a crucial foundation for advances in understanding and modeling of the climate system. To reach the scientific goals outlined above, the ASR program will make detailed measurements of the aerosol-cloud-precipitation continuum, the processes that define this continuum, and the radiative and dynamical environment in which it exists. A major element of these activities is to derive the relevant geophysical properties from these measurements, an activity that will itself require a substantial research effort. In coordination with these observational efforts is the *process modeling* component of the ASR program, which will incorporate the geophysical data sets into modeling frameworks and analyses at various scales to develop a more comprehensive understanding, and an improved model representation, of the climatically important atmospheric processes. By pursuing these complementary goals of measuring and modeling simultaneously, ASR will achieve more efficient advancement in both areas.

The methods and tools that will be utilized to implement the ASR program's scientific objectives are outlined in this section for both observational and process modeling activities. These two sets of activities are intimately linked. Model studies and uncertainties establish observational priorities by specifically identifying the geophysical parameters and processes that must be better understood. In turn, observational data sets are used to initialize, constrain, evaluate, and ultimately develop model parameterizations. One of the ASR program's characteristics is its efforts to enable more effective feedback between observational work and model studies.

The ASR program's observational and modeling approach is to:

- Determine the essential characteristics of the coordinated laboratory and field measurements necessary to understand aerosol and cloud life cycles and aerosol-cloud-precipitation-radiation interactions (e.g., variables measured, measurement accuracy, sampling strategies).
- Develop research strategies to create the integrated data products necessary to improve the understanding of aerosol-cloud-precipitation-radiation interactions (e.g., retrieval development, uncertainty analysis, data product collocation, quality control).
- Utilize the integrated data products to evaluate, and ultimately improve, the parameterization of aerosol-cloud-precipitation-radiation processes in models over a range of scales (e.g., what data products are needed, what metrics are used to evaluate model improvement).
- Evaluate the trade-off between the minimum level of model complexity and model accuracy required to represent the range of atmospheric conditions that determine climatically relevant aerosol and cloud properties and aerosol-cloud-precipitation-radiation interactions.

3.1 Observational Methods and Tools

ASR is an observation-based program, whereby measurement data are used to derive new insights into atmospheric processes and to critically evaluate the results from numerical models. The ARM Facility has a wide range of in situ and remote sensing instrumentation that can be used to characterize the atmospheric thermodynamic structure, cloud properties, aerosol properties, and the radiative environment. New observational tools acquired as part of the ARRA of 2009 greatly enhance the capabilities of the

ARM Facility, providing the capability for developing new insights into cloud and aerosol properties, dynamics, and precipitation that were not possible before. The ARM Facility enhanced observational suite is now able to more completely observe properties and processes along the entire range of the aerosol-cloud-precipitation continuum, and will provide the data needed to evaluate the range of processes outlined in Section 2. ASR and the ARM Facility will be tightly coupled, with ARM observations used in ASR analyses and ASR providing guidance on the essential characteristics that the ARM data sets need to have.

Laboratory studies, informed by short-term intensive field campaigns, are necessary to support and supplement the long-term ARM Facility observations. Laboratory studies are essential to unraveling processes involving chemical reactions and composition-dependent aerosol microphysical properties. Examples include investigating the effects of size-dependent composition and mixing state on CCN, IN, and particle optical properties; reactions of organic precursor gases; and the mechanisms of new particle and SOA formation. These studies are vital for the development and testing of next-generation, field-deployable measurement technology and for measurements-based investigations of key atmospheric processes under controlled conditions. The design and optimization of cloud and aerosol microphysical instrumentation is key to developing insight and understanding of atmospheric processes. Examples for clouds include the development of new and more compact CCN and IN spectrometers, cloud particle sampling probes that alleviate problems associated with shattering and bouncing of ice particles at probe inlets, and in situ methods to measure the optical properties of hydrometeors within clouds. Examples for aerosols include instruments capable of measuring particle size, shape and speciation, ideally on a single-particle basis and at frequencies suitable for aircraft sampling, and extending the size range of field-deployable particle measurements.

Typically, the measurement made by any instrument (e.g., a voltage) is not what is ultimately desired. Rather, the needed geophysical variable must either be derived or retrieved from the raw measurements. This retrieval problem is often ill conditioned, and thus benefits from the synergistic use of multiple coordinated sensors that can provide a more accurate measure of the geophysical variable. It is critical that uncertainties in the raw observations and a priori information used in the retrieval process be propagated through physical models to provide uncertainties in the retrieved variable. Accurate characterization of uncertainties in all geophysical variables derived from ARM Facility data is a high priority of the ASR program. ASR will develop well-characterized data products with uncertainties to allow for clarity in research and the most efficient means for evaluating model simulations.

3.1.1 Aerosol Observations

Understanding the life cycle of aerosols and characterizing their optical, chemical, and microphysical properties requires both *in situ and remote sensing observations*. In situ measurements, which may be made continuously at surface stations, yield essential microphysical, chemical, and optical properties of aerosols and permit inference of governing processes. Such measurements readily allow local closure experiments relating optical and CCN and IN properties to size distributed composition, and permit aerosol properties to be related to measures of aerosol processing such as photochemical age and dilution. However, in situ observations made at the surface are generally not representative of aerosol in the full atmospheric column. Consequently aircraft measurements, although inevitably limited in space and time, are essential to understanding the three-dimensional distribution of aerosols, their properties affecting radiation and clouds, and their evolution. A major challenge to ASR is extending the capabilities and

coverage of in situ observations through developing and applying means of using ground-based remote-sensing observations, suitable for long-term analyses, to provide the information needed to determine aerosol properties and aerosol influences on clouds and radiation. The addition of the Mobile Aerosol Observing System to the ARM Facility, an aerosol observing system at the Darwin ARM site, aerosol instrumentation for the second Mobile Facility, and new aerosol instruments for the Aerial Facility will be key to studying aerosol life cycle questions that are a focus of ASR.

New particle formation, which affects the number concentration of particles, is a key process in aerosol dynamics that governs size distributed composition and in turn the aerosol optical and cloud nucleating properties. New particle formation needs to be better characterized under a variety of controlling environmental conditions, especially the role of organics; this need is driving development of instrumentation to extend measurements of particle composition to smaller and smaller particle size. Also important is the need to identify gas phase precursors and determine their concentrations and the dependence of new particle formation rates on these substances. The latter need is driving the application of mass spectrometry using gentle ionization techniques such as chemical ionization to preserve the molecular structure of gas-phase precursors. Application of these new techniques in laboratory studies and field measurements will undoubtedly help to bridge the key gap in understanding of processes responsible for stable new particles in the size range between molecular clusters and particles of current minimum detection size (about 3 nm), thereby allowing confident representation of these processes in models.

As particles grow their ability to serve as CCN or IN depends strongly on the size and composition of the particle. ASR considers it critical to understand the different chemical and physical characteristics that influence the *formation of liquid and/or ice on an aerosol particle* and to improve techniques to characterize CCN and IN as a function of supersaturation. Laboratory studies under controlled conditions and field studies in ambient conditions are both necessary. Techniques are needed to permit determination of the size and composition of cloud particle nuclei as well as to distinguish between ice and droplet nuclei in order to characterize the properties of the small fraction of particles that are ice nuclei.

Models typically simulate far less *secondary organic aerosol (SOA)* than is observed in the atmosphere. Laboratory and field studies of both gas- and aqueous-phase SOA formation and investigations on how SOA particles mix with other aerosol types are needed. The aging of these particles due to both the condensation of secondary material on the particle and coagulation with other particles is widely considered to impact their ability to serve as CCN and to absorb radiant energy. These studies require a wide range of observational tools to characterize the gaseous precursor concentrations, aerosol size distribution, chemical composition, and absorptive properties; many of these tools are already operated within or are being acquired by the ARM Facility and techniques are being developed to better utilize these observations to gain a better understanding of the physical processes along the SOA lifecycle.

Ultimately, determining the full impact of aerosols throughout their lifecycle on clouds, precipitation, and the radiation budget requires knowledge of the spatial distributions of *aerosol microphysical and optical properties* over time. The vertical distribution of light-absorbing aerosol impacts the diabatic heating rate profile, especially in cloud-free scenes, and hence changes the thermodynamic structure of the atmosphere. Aerosol optical depth is routinely measured at ARM Facility fixed and mobile sites using shortwave ground-based radiometry, but is limited to column averages in cloud-free conditions, whereas active remote sensing by lidar is used to determine the vertical distribution of aerosol under clear and

cloudy conditions. The Doppler and Raman lidars to be acquired by the ARM Facility will further understanding in aerosol-cloud-precipitation-radiation processes with the capability to determine both aerosol vertical profiles and vertical air velocity. Observations of the concentration of CCN and the vertical velocity near cloud base, together with accurate measurements of cloud properties and radiative fluxes, will be used to develop and test representations describing the impact of aerosols on cloud microphysical properties. Aerosol absorption, its spectral dependence, and its dependence on relative humidity are difficult to measure with ground-based remote sensors. ASR will take advantage of recent advances in aircraft instrumentation to study how absorbing aerosol is vertically distributed in different geographical locations and its consequent impacts on vertically distributed radiative fluxes and heating. Improved remotely sensed measurements of total column aerosol absorption, especially in low aerosol optical depth conditions, will also be pursued.

3.1.2 Cloud Observations

Clouds are difficult to model because of their complexity and wide range of pertinent scales. They lie at the intersection of vapor, liquid, and ice and are subject to many microphysical processes that convert water from one of these phases to another. Clouds and cloud system life times range from minutes to weeks, their spatial scales range from meters to hundreds of kilometers, and their important processes can occur at the microscale to the synoptic scale. Thus, a wide range of in situ and remote sensor observations is needed to improve understanding of the cloud life cycle.

To first order, the most important variable needed to understand the cloud life cycle and its impact on the radiative and hydrological cycles is the *amount of condensed cloud water*, both instantaneously and statistically over time. The ASR program will emphasize the measurement of liquid water path (LWP) and ice water path (IWP) using vertically pointing instrumentation, with a specific focus on improving and characterizing the retrieval accuracy. Of particular importance are improved retrievals of LWP in thin clouds, which have a very sensitive interaction with atmospheric radiation, utilizing spectral infrared measurements and newly acquired microwave radiometer channels, among other strategies. In addition, the three-dimensional distribution of liquid and ice water content (LWC and IWC, respectively) as a function of time, and especially statistics that describe these distributions under a variety of synoptic conditions, are essential. Characterizing these important cloud properties will require a coordinated effort to optimally combine information from a variety of instruments, new and old, including infrared and solar spectrometers, lidar, scanning microwave radiometer, scanning radar, and aircraft in situ sensors.

Serious difficulties are encountered when attempting to quantify IWC and IWP. Remote sensor-based retrievals often rely on uncertain assumptions about ice crystal habit (shape) and effective density. Moreover, in situ ice measurements have historically suffered from crystals shattering on probe inlets, resulting in large biases in the crystal concentration as a function of particle size, especially for crystals smaller than 50 μm . Both of these issues present challenges for interpreting past observations of IWC and IWP, which were used to construct model parameterizations. These difficulties support the need for new and improved methods. ASR is poised to better observe the ice crystal habit and effective density using improved aircraft probes that reduce shattering artifacts, new measurements from scanning radars that will be validated against aircraft in situ crystal imaging probes, and advanced lidars that are able to measure the ice cloud extinction profile directly.

In addition to bulk cloud properties, the *cloud particle size distribution* and its dispersion (width) are important controlling factors in many processes, including radiative transfer through clouds, interactions of cloud particles with aerosols, and precipitation formation. Thus, improved techniques to characterize the cloud particle size distribution will contribute to a better understanding of crucial linkages in the aerosol-cloud-precipitation continuum. Some remote sensing techniques are able to retrieve cloud particle effective radius, an important characteristic of the full size distribution; however, these measurements struggle to explicitly describe the complete size distribution. While fully characterizing the particle size distribution in all conditions will continue to be difficult, targeted aircraft campaigns using sensors that minimize crystal shattering will be the key to future improvements and may help to unlock the size distribution information contained in cloud radar Doppler spectra. In coordination with cloud measurements, in situ observations of cloud condensation nuclei spectra will be made below and above cloud layers. These measurements, augmented by fast turbulence gust-probe measurements of vertical velocity, will provide valuable data sets for testing theories and developing parameterizations of processes such as cloud droplet and drizzle formation. ASR will also pursue synergetic techniques that combine multiple observations with different sensitivities to the particle size distribution to better characterize the size distribution in a wider range of conditions.

Mixed-phase clouds, where water exists in all three phases simultaneously, are extremely challenging to model. The long-term and campaign data sets collected at the ARM North Slope of Alaska site have greatly improved knowledge of mixed-phase cloud processes, yet there is still much to learn. In these clouds, the liquid water is supercooled, making in situ observations more difficult due to aircraft and probe icing effects. Many remote sensing techniques are sensitive more to one phase of cloud particles than the other (liquid vs. ice), limiting the information available from any one instrument. Furthermore, as in all clouds, accurate characterization of the ambient relative humidity is needed to better understand the interaction between vapor and the condensed phases. ASR will therefore pursue mixed-phase cloud characterization by using synergetic measurements and retrieval methods based on unique mixed-phase signatures from instruments such as depolarization lidars, cloud radar Doppler spectra, infrared spectrometers, and aircraft in situ probes capable of measuring across the full range of both liquid and ice particle sizes.

3.1.3 Precipitation Observations

Like clouds, precipitation occurs on a variety of scales and intensities, which are determined by the environmental conditions. The ARM Facility's new three-dimensional (3D) observing capabilities, based on scanning radiometers and precipitation radars, provide a detailed view of precipitation on a range of scales and will allow for the processes associated with precipitation to be studied much more extensively than is currently possible.

A primary initial focus for ASR is to obtain 3D measurements of the *precipitation field* around the ARM Facility sites using the new scanning precipitation radars. This includes liquid and ice precipitation in both convective and stratiform conditions. The separation of total condensed water path into cloud and precipitation components is important for model validation and to understand the processes linking clouds and precipitation. ASR will place an increased emphasis on developing the multi-sensor techniques that are needed to observe and understand these precipitation processes. A related and important geophysical variable that is needed for model evaluation is the *latent heating rate profile*, and methods are needed to derive this variable from the precipitation observations. For both of these cases, it is imperative for ASR

to design radar scanning strategies that can best capture the 3D distribution of precipitation while also placing these measurements within the context of historical and ongoing vertically pointing measurements.

Microphysical processes are important to the onset, intensity, and duration of precipitation events. Studies have shown, for example, that *dispersion (width) of the droplet size distribution* is one of the main controlling factors in drizzle and rain formation. Additionally, uncertainties in collision and coalescence processes and the role of aerosol entrainment into precipitating clouds lead to large uncertainties in numerical model simulations. ARM Facility observations will be used to investigate the factors that control droplet dispersion and the subsequent growth of precipitation-size hydrometeors (e.g., entrainment, updraft velocity, turbulence, and relative humidity). Due to the complexities associated with precipitation formation, ASR will construct a hierarchy of observational strategies including aircraft missions examining specific processes that impact precipitation such as entrainment; ground-based scanning radars to understand the broader spatial organization of precipitation systems; and vertically pointing Doppler lidars and radars to target vertical velocity and turbulence information.

3.1.4 Radiation Observations

Major advancements in the understanding of radiative transfer, especially in cloud-free atmospheres, have been made by ARM, yet several questions remain unanswered and many new applications can be addressed using the ARM Facility's updated and new radiometry. *Spectrally resolved radiance and flux observations* in the solar and infrared spectral regions are crucial to the understanding of both clear-sky and cloudy radiative processes. An extensive data set (~15 years) of downwelling longwave spectral radiance has been collected at many locations, and the recent increase in the temporal resolution in this data by an order of magnitude is enabling more accurate cloud property retrievals from these data. These observations have been used to improve detailed clear sky infrared radiative transfer models, as well as the parameterization of infrared radiative transfer used in climate models. Additionally, aerosol properties, including composition, have been retrieved from spectral infrared observations in African dust cases, although, with more work is needed to fully mine the infrared spectra to provide more information on aerosol types and atmospheric properties.

The shortwave spectral data set acquired by ARM spans less than two years at a single site, as opposed to the spectral infrared data sets that span over a decade at multiple sites. Due to the resulting paucity of spectrally resolved observations in the visible and near-infrared at the ARM Facility sites and the limited climatological sampling, there remain many uncertainties associated with *spectral shortwave radiative transfer*, and in particular in the magnitude of the absorption or solar radiation by clouds and aerosols. The new spectral shortwave measurements at the ARM Facility sites, made possible by the ARRA funds, will be used to investigate the accuracy of gaseous, liquid, and ice absorption models in the visible and near-infrared portions of the spectrum. The new observations will ensure that the spectral absorption by water vapor, aerosols, and clouds are properly treated in climate models. These studies require that the spectral surface albedo, especially in the near-infrared, be well characterized. These new spectral observations will, in particular, be used to investigate the radiative properties of the humidified aerosol that typically surrounds cumulus clouds, which is one component of the aerosol-cloud continuum.

Cloud property retrievals, especially of optical depth from passive radiometers, will continue to be improved and new techniques will be developed that integrate spectral radiance observations with those

from other remote sensors to retrieve additional information about cloud microphysical properties (e.g., ice crystal habit, droplet dispersion in thin liquid clouds, etc.) that are not possible using a single instrument. The addition of the new 90 GHz channel in the ARM Facility's new 3-channel microwave radiometers provides increased sensitivity to LWP, which is especially important for accurately characterizing clouds with LWP values less than 100 g/m^2 that occur very frequently in nature. The accuracy of the forward microwave absorption model at 90 GHz, and in particular the temperature sensitivity of the liquid water absorption, will be tested and improved.

From a radiative point of view, the most important cloud property is the optical depth. Recent efforts by ARM investigators have greatly expanded the complement of tools that can retrieve optical depth, but most of these methods do not provide information on the *vertical distribution of the optical depth* (i.e., the extinction profile). Advanced lidars, such as the new Raman lidar and the high-spectral-resolution lidars that are the result of the ARRA procurement, are able to directly measure the extinction profiles in cases where the optical depth is less than approximately 3. Combining measurements of the extinction profile of these clouds with other observations allows information such as the profiles of water content and the effective cloud particle size to be retrieved more accurately. These data products, which currently require development and validation, will greatly aid investigations of the radiative properties of cirrus where these advanced lidars are deployed. In particular, these lidar observations, together with other remote sensing observations, will be used to investigate the importance of small ice crystals in the cirrus particle size distribution and the impact of these small crystals on the radiative fields and lifetime of the cloud.

The radiative influence of *ozone* is climatically important in both the solar and thermal spectral regions, as it is one of the most important greenhouse gases after carbon dioxide. Heretofore, the ARM Facility has relied on once-a-day NASA satellite overpasses to provide column ozone measurements. Since the tropospheric ozone concentration can change significantly in only a few hours, daily measurements are inadequate for precise testing of radiative transfer models. Also, discriminating the tropospheric component (small) from stratospheric component (large) of the total ozone amount is crucial; it is the tropospheric component that varies the most due to anthropogenic influences and it is nearly impossible to retrieve accurately from satellites. ASR should therefore pursue methods to measure tropospheric ozone concentration from the surface.

3.1.5 Dynamics and Thermodynamics Observations

Clouds and aerosols evolve in an environment where many factors can influence the various processes that affect their lifetimes. Thus, an improved characterization of this environment is crucial.

Water vapor, and in particular *relative humidity*, is a key factor that influences many processes associated with clouds, aerosol, precipitation, and radiation. Thus, ASR needs to produce water vapor profiles with sufficient accuracy and temporal and vertical resolutions to resolve variability in this important parameter. Water vapor is currently measured in situ by radiosondes and aircraft; remote sensing methods include direct measurements of water vapor by Raman lidar and retrievals from spectrally-resolved infrared observations. The column-integrated water vapor, also called precipitable water vapor (PWV), is a strong constraint and is used to calibrate the profiles derived from remote sensors. Continued effort must be placed on improving PWV retrievals in very dry and moist conditions ($< 1 \text{ cm}$ and $> 4 \text{ cm}$, respectively). ASR will continue to characterize and improve radiosonde water vapor measurements, and will place a large emphasis on the retrieval of water vapor from the ground-based remote sensors, especially in cloudy

environments. In particular, ASR will focus on accurately observing humidity in the upper troposphere, and to improve the accuracy and vertical/temporal resolution of humidity measurements in the boundary layer throughout the diurnal cycle, especially near the top of the boundary layer where clouds often form.

The lifetime of cloud particles is strongly tied to ambient conditions and *cloud-scale dynamics*. Water vapor condenses to liquid or ice only when saturation conditions occur, which typically coincide with updrafts. Entrainment of drier ambient air or more CCN into the cloud can have a large impact on the microphysical properties of the cloud and hence its lifetime and radiative properties. The new observational capabilities of the ARM Facility allow these cloud-scale dynamical conditions to be better measured. Doppler spectra from the cloud and precipitation radars provide a wealth of radial velocity information on hydrometeor movement; however, new techniques are needed to account for particle fall speed and derive 3D maps of vertical velocity. The addition of Doppler lidars at several of the ARM Facility sites provides another measure of vertical velocity that complements the Doppler radar observations due to the different weighting by particle size. ASR will develop algorithms that utilize both of these data sets to determine the cloud scale air motion and fall speed, and to investigate turbulent processes within clouds. In addition, data from the Doppler lidars and high-resolution water vapor observations from the Raman lidar will be used to develop statistics on the turbulent structure in the boundary layer in cloud-free scenes; these observations will be used to relate turbulent structure with other variables (e.g., shear at the top of the boundary layer, surface heat fluxes) to develop and improve parameterizations of boundary layer turbulence.

The characterization of the surface layer is essential for ASR modeling efforts. *Surface latent and sensible heat fluxes*, which are important boundary conditions, can be quite variable over an inhomogeneous surface. The program will continue to emphasize the measurement of these surface fluxes and to use these fluxes to initialize and evaluate models. Each site faces unique challenges associated with this activity, especially the mobile facility sites that often lack some of the additional infrastructure associated with the fixed ARM Facility sites.

3.2 Process Modeling Methods and Tools

An overarching goal of ASR is to narrow the uncertainties in the model-based prediction of future climate for a given emission scenario, specifically the uncertainties related to the representation of aerosol and cloud processes. The previous sections summarize the specific aerosol, cloud, and precipitation processes targeted for improvement, as well as the key measurement strategies that will be implemented to gather the required data. This section outlines the final step in an iterative loop, wherein physical and chemical understanding of the pertinent processes is implemented in models and model performance rigorously evaluated to ensure that these processes are properly represented. While one end goal of ASR is to improve GCMs, the tremendous difference between the resolution of GCMs (hundreds of km in horizontal resolution, for instance) and the resolution required to resolve virtually all relevant physical processes (generally much less than 1 km for cloud processes and their interaction with aerosols) suggests that using a hierarchy of models with decreasing domain sizes is the most effective strategy to determine whether interdependent processes are properly understood and evaluated using the ground truth provided by field and laboratory measurements. This strategy is illustrated in Figure 6.

3.2.1 Process Model Development

There is a continuing need to develop models in order to incorporate (1) the latest findings on aerosol and cloud life cycle processes and (2) new approaches for representing these processes numerically. ASR activities will span the full range from process-level parameterization development at the box-model level to the incorporation of new parameterizations and observational constraints and testing these at the GCM scales.

As described in Section 2.1, modeling of the *aerosol life cycle* requires representation of transport, emissions, new particle and secondary aerosol formation, aging, mixing state, optical properties, and removal. To maintain their relevance, aerosol life cycle models need to evolve as measurements evolve; however, comprehensive aerosol models are computationally expensive and thus some simplifications via parameterizations are needed. For example, new particle-resolved size/composition measurements, which are capable of resolving general mixing states, pose a special challenge. Whereas some detailed process and regional models can afford to divide aerosol size distributions explicitly into size range “bins” and represent properties independently in each bin, even these require that all particles in the same size bin have the same composition (assumption of internal mixing) and are thus inadequate for treating the general mixing states of a multi-component aerosol. Simulations capable of separately tracking the life histories of a million or more particles using Monte Carlo methods, while excellent for testing approximations at the box model level, are far too computationally demanding for application in larger scale models such as a GCM. Integral to all models is the representation of the aerosol particle distribution function. Global models must select a minimum number of tracers (transported scalars) to achieve this representation. Tracers can be individual bin populations, modal properties (e.g. number and mass of a prescribed particle distribution function), or selected statistical moments of a multivariate generally mixed population. ASR will support research in all of these approaches in order to most rapidly accelerate understanding of aerosol processes using smaller-scale models, as well as to roll up such knowledge into improved regional and global-scale models.

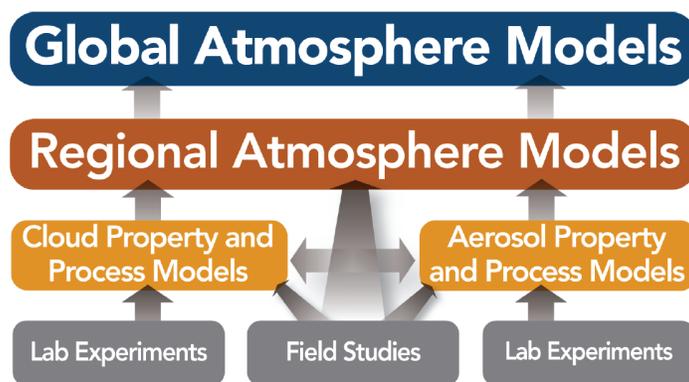


Figure 6. A strategy for improving the representation of cloud and aerosol properties and processes in climate models (adapted from Ghan and Schwartz 2007). Laboratory experiments provide data used to improve understanding of isolated cloud and aerosol processes and constrain process models. Field studies (both long-term and intensive) provide data for testing process models. The suite of all process models governing the life cycles of clouds and aerosols are applied to regional atmosphere models (large eddy simulation, cloud-resolving, single-column, and mesoscale), and these integrated models are also evaluated using data from the field studies. The process models are then applied to global models, which are evaluated with the field study data and with satellite data.

In the cloud life cycle (as in the aerosol life cycle), a wide range of dominant physical processes occur on unresolved scales (e.g., entrainment of clear air) and these must be approximated or parameterized in model types ranging from large-eddy simulation (often 20-100 m horizontal resolution) to cloud-resolving model (often 4-10 km horizontal resolution) to GCM (generally >100 km resolution). Improving *cloud microphysical parameterizations* is especially important to assess aerosol indirect effects. “Bulk schemes” diagnose cloud liquid and ice mass, but more simply specify the effective cloud particle size for radiative purposes and include only limited treatment of complex mixed-phase processes. More advanced schemes include the computation of at least cloud particle number concentration and mass (i.e., a “two-moment scheme” because these are the 0th and 3rd moments of the particle size distribution, respectively). These enable the mean cloud particle size to evolve in response to aerosols, which is necessary for representing aerosol indirect effects on radiative transfer and precipitation formation, as well as chemistry–cloud–aerosol interactions such as wet deposition and scavenging. Two-moment schemes have been implemented in both regional-scale models and GCMs, but much work remains to be done, as in the representation of subgrid variability. Given the importance of multi-moment schemes to representing aerosol–cloud interactions in GCMs, ASR will continue to support their further development and evaluation. Similar to representation of aerosols, so-called “bin” or sectional models also exist to represent cloud particle size distributions in the most generalized and detailed fashion for study of specific complex processes in smaller-scale models. However, the detailed microphysical data that must be input to bin models (e.g., efficiency of riming for specific pairs of hail and droplet sizes as a function of atmospheric state) are often not well constrained by laboratory studies to date and are also not well evaluated using field experiments. ASR is well positioned to identify the key uncertainties in bin models and resolve them with targeted laboratory and field studies, thereby accelerating bin model use to conclusively resolve rain and ice formation process questions, among others.

There are many physical processes that influence the cloud life cycle must be better understood and represented in numerical models. A key GCM subgrid-scale process is *atmospheric convection*, as it controls the formation of convective precipitation, provides latent heat that drives atmospheric circulations, and transports water vapor, aerosols, and chemical species vertically. The parameterization of atmospheric convection has proven difficult as there are many characteristics of convection that must be understood and represented, such as when and where convection occurs, how much mass is transported between vertical levels (including the critical issue of how much environmental air is entrained into a convective updraft plume), the vertical velocity of air in the updraft plumes, how much precipitation evaporates before reaching the surface, whether such evaporation creates downdrafts that cool and dry the boundary layer, and the degree to which convective organization at the mesoscale affects the lifetime of convective systems. Turbulent mixing is another physical process that requires improvement in GCMs. Some specific challenges with representing mixing in models include the rate of entrainment of the overlying air into the boundary layer, whether mixing between clear and cloudy air is homogeneous (cloud particle number conserving) or inhomogeneous (non-conserving), and the fraction of a grid-box with cloud and its vertical overlap. ASR can make key contributions to the development of improved convection and mixing parameterizations through its extensive observations of physical properties like vertical velocity that are retrieved from scanning cloud and precipitation radars.

An especially important effort dedicated to advancing cloud modeling is the ongoing development of *large-scale forcing data sets* for ARM Facility measurement sites. Because the essential activity of cloud parameterization involves relating mean grid-scale cloud and atmospheric state evolution to initial environmental conditions, it is crucial to sufficiently characterize the large-scale environment for many modeling studies. However, many important large-scale variables (such as mean vertical wind) are

currently impossible to measure directly, whereas others (such as how much net moistening occurs at a given level of the atmosphere over a wide area) are prohibitively expensive to measure routinely. ASR and the ARM Facility have pursued several approaches to this problem. For intensive field campaigns, where detailed data is most important, the ARM Facility incurs the significant expense of funding frequent balloon releases over a widely-dispersed array of sounding sites; this is currently the most complete method of characterizing the large-scale dynamical and thermodynamical environment around storm systems, for instance. When combined with wide-area precipitation data in a theoretical framework, large-scale mean vertical wind can be calculated. For routine operations, the ARM Facility continues to fine-tune methods for integrating wide-area precipitation radar data at the local sites (and other ancillary data sources that depend upon availability at each site) with the atmospheric state derived from reanalysis model runs that in turn assimilate routine ground-based and satellite data nationally and internationally. These integrated forcing data sets provide all of the important environmental variables needed to run a model over a limited size domain. Model calculations can then be compared with aerosol and cloud properties measured at a central site or averaged over the domain. The ASR program will continue to support this well-developed method of forcing models, in addition to actively developing new methods.

Because a goal of ASR is to develop models of aerosol and cloud processes that can be applied to climate models, it is vital to evaluate these process models under real-world conditions for a variety of regions and seasons. Regional atmosphere models representing the integrated set of all processes that determine aerosol and cloud properties provide *flexible frameworks* for developing and testing modules of individual processes. Regional models include cloud-resolving models, single-column models derived from a global atmosphere model, and chemical transport models. Integration of all aerosol and cloud processes is more straightforward if the effort is focused on a single modeling framework that can operate in any of these modes. Such frameworks already exist and are being extended to appropriately include land, ocean, and sea-ice surface processes. Ideally all investigators would adopt common frameworks so that the performance of candidate parameterizations of isolated processes can be compared with other parameterizations for the same conditions and parameterizations of other processes. A variety of cloud and aerosol modules can be supported within such a framework, with more detailed modules applied at fine spatial resolution serving as benchmarks for comparison with coarser resolution simulations using simplified modules designed for GCMs. The goal is to determine the minimum level of complexity needed to accurately represent climatically important aerosol and cloud processes in models. Data from field experiments are particularly well suited for evaluating cloud and aerosol processes simulated by regional models. These models can also be used to guide experiment design, determining optimal locations for surface sites and sampling strategies for aircraft flights.

A particular challenge for parameterization of atmospheric processes is a scalable modeling framework, which essentially constitutes *portability of parameterizations to models that operate on different space and time grid scales* (e.g., model simulations over the vastly different spatial scales depicted in Figure 4). Often, tuning of parameters is used to preserve model performance as grid cell size diverges from process-level scales; this tuning is, by necessity, specific to model scale. Frequently the tuning of parameters, especially parameters that have direct physical meaning, is both arbitrary and incompatible with any scalable modeling framework. Consider, for example, the critical droplet radius r_c beyond which cloud droplets enter the collection regime and rapidly grow to fall velocity size. Measurements have shown that drizzle often takes place within localized regions of cloud that are sub-grid to cloud resolving models. Averaging cloud liquid water content L over the full model grid cell can easily result in L values too small to support drizzle, leading many modelers to tune r_c to smaller values in order to match

observations. The preceding is but one example of what is a pervasive problem for atmospheric process modeling on the global scale. Ideally one would like to have scale-invariant parameterizations, but the route forward to their development is not clear-cut and will likely require bold and creative innovation. One approach is to use probability distribution functions (pdfs) based on ARM Facility cloud observation data sets, for example, to map from the local microphysical scale to a probabilistic representation of sub-grid cloud properties at the large eddy, cloud resolving, and GCM scales. Another approach is to let the models themselves generate the statistics needed for the probabilistic representation of sub-grid properties, for instance either using ensemble model runs, to generate sufficient statistics for generating sub-grid pdfs, or through interpolation. A third approach is to adapt image-processing methods based on fractal interpolation and/or iterated functions systems, which are techniques used to provide “texture” at the sub-pixel scale of lower resolution images. While the best methods to achieve scale-invariant parameterization remain to be determined, this challenging problem is of such clear and vital importance that it will be an important focus of ASR model development.

Finally, advances in cloud parameterization need to be fully integrated with advances in aerosol parameterization. The centerpiece of this progress will be an emphasis on multi-moment aerosol and cloud schemes for GCMs. Success will depend upon understanding and parameterizing the processes that describe aerosol and cloud properties and upon linking aerosol and cloud life cycles.

3.2.2 Model Evaluation

ASR aims to bring *convergence to aerosol and cloud parameterizations* similar to that achieved for radiation parameterizations, which was a significant accomplishment of the ARM Program. This desire for convergence is an aggressive goal because complexities associated with cloud and aerosol life cycles and the associated processes are very high relative to those associated with radiative processes. In order to guide convergence of aerosol and cloud models, ASR will promote the development and improvement of integrated data sets, with which aerosol and cloud parameterizations and model fidelity for various atmospheric processes can be evaluated across the full range of model scales. ASR will also iteratively tailor ARM Facility data collection goals to optimize the simultaneous development of model diagnostics alongside integrated data sets.

Perhaps the single most important meeting point of measurements and models in the ASR program is at the level of *integrated data products*. These consist of a growing set of data streams that select one or more “best-estimate” of geophysical variables from a set of data streams from one or more instruments; such derived data streams are referred to within the program as “value-added products”. The most widely used value-added product within the ARM Facility data set is the Active Remotely Sensed Clouds Locations product, which combines data from active remote sensors to produce an objective determination of hydrometeor height distributions, radar reflectivities, Doppler velocities, and Doppler spectral widths. A second, recently completed value-added product that is rapidly receiving wide use in the modeling community is the Climate Modeling Best Estimate, which is specifically tailored for use in evaluation of regional and global climate models. This data product contains hourly average best estimates of ten selected observational measurements (e.g., cloud fraction, surface radiation fluxes, total cloud cover, liquid water path, and precipitable water vapor) at the ARM Facility mid-latitude site. Also underway is the development of a new Radiatively Important Products Best Estimate data product that will include collocated radiation, aerosol, and cloud variables at high temporal and spatial resolution. The task of collating measurements in a systematic way would be extremely time-consuming if done individually by each modeling group accessing ASR data; efficient use of ASR data demands that this

process of providing polished end products be pursued. By far the most valuable data for model evaluation and improvement consist of data sets that contain more than one collocated variable.

At the global scale, it has long been desired to establish a set of metrics by which climate models can be evaluated, but progress toward this goal has been elusive. The meteorology community has well-established metrics for weather forecasts that are based on subsequent observations of the quantities they predict, thus ensuring that the metrics are relevant to the science goal of the models. *An identical metrics-based approach cannot be taken for climate models* for several reasons. First, climate models are asked to forecast many different things (forcings, feedbacks, regional impacts), and each of these may require a different set of metrics. Second, climate models forecast changes on longer time scales than those for which observations are available, and thus, unlike numerical weather prediction models, the utility of any metric cannot be definitively established. Third, many of the things that are most important for climate models to forecast well are too poorly observed, or observed over too short a time scale, to act as effective constraints. Previous attempts to define metrics have relied primarily on spatial distributions of mean quantities rather than temporal variations. One difficulty with this is that a simpler parameterization with more free parameters to tune may outperform a parameterization that tries to represent more of the actual physical interactions that may be important in a climate change. Finally, studies of variability of climate models have often identified a subset of models that perform the best, but are often unable to identify a parameterization approach that the successful models have in common, because the cloud and convection parameterizations interact with all other aspects of a GCM, in different ways in different models.

Given this complexity, ASR will focus on the specific task of defining and applying *observation-based diagnostics* that measure the fidelity with which models simulate aerosol and cloud processes. For representation of cloud properties, one approach will be to make use of the growing library of GEWEX Cloud System Study (GCSS) program case studies, several of which have been led by ARM Program scientists and are based upon ARM Facility observations. ASR and the ARM Facility will support major new field campaigns that will be suitable for expanding the number of well-observed case studies. Judiciously chosen case studies will continue to be used to evaluate specific cloud parameterizations in a more controlled setting, such as using regional models or single column model versions of GCMs that isolate grid-scale physics from dynamics in the larger GCM. Different parameterization approaches are sometimes incompatible with one other or with other aspects of the model in which they are embedded. Therefore, ASR will pursue a strategy of diagnosing how the different parameterizations of cloud and aerosol processes interact within the model to ensure consistency with the observations for that case.

An important focus area for ASR model evaluation efforts will be the development and application of *instrument simulators*. Most remote-sensing instruments do not directly measure modeled variables (such as mass concentration of ice particles in clouds) but instead quantitatively infer the variable from measured radiative parameters. However, depending upon model type, model outputs may be sufficient to directly calculate quantities that are actually measured (e.g. visible and infrared radiances, radar reflectivity, and lidar backscatter signals). The modeling community has developed software packages known as “simulators” for this purpose. Simulators can have the advantage of avoiding some types of retrieval error that otherwise add to the error already inherent in the raw observations. The ISCCP Simulator developed by ARM scientists, which was the first such simulator, is now routinely used by many modeling groups. Simulators have been invaluable for comparing models with each other and to observations from remote sensing instruments, for pointing out systematic biases of climate models, and for analyzing cloud feedbacks. Development of simulators for existing and new ARM instruments will be a high priority for ASR.

4.0 Future Directions

This document outlines an aggressive and comprehensive plan that will greatly advance the understanding of the myriad processes that comprise the aerosol and cloud life cycles as well as the interactions among these processes. A tight coupling between ARM Facility observational capabilities and ASR scientific expertise is critical to the success of this research enterprise. However, to most effectively utilize the ARM Facility observations to advance the representation of atmospheric processes within numerical models as outlined in this plan, substantially larger budgets are required for both the ASR program and the ARM Facility. Specifically, expanded efforts are needed to produce integrated data sets that utilize the new observational tools provided by ARRA and to perform the analyses necessary to link, broaden, and integrate the research foci of the former ARM and ASP Programs. Development of integrated data sets and incorporation of these data sets into observational products and model improvements is the highest priority for growth within the program; however, if support for further activities becomes available, the activities outlined below will further contribute toward the ultimate goal of process-level understanding of the aerosol and cloud life cycles, their interactions, and reducing uncertainty in climate simulations and projections.

ASR will make large advances towards understanding the atmospheric processes that are needed to improve the accuracy of climate models with the wealth of data provided by the combination of ARM Facility fixed sites, mobile facility deployments, and deployments of the aerial facility. However, as indicated earlier, some atmospheric processes are more important and more readily observable, relative to others, in certain climatic regions and thus it is important to investigate the details of these processes in these regions. Through longstanding discussions, ASR has a plan to target specific regions and processes in the future if given the opportunity to do so.

There is a general consensus in the climate modeling community that the largest differences among predictions of future mean surface temperature from different climate models occur in the regions that are most heavily influenced by stratocumulus clouds on the west coasts of the various continents (Bony and Dufresne 2005, Figure 7). The importance of stratocumulus regions has been known for many years, and was one of the primary motivations for deploying the ARM Mobile Facility to the Azores for 18 months in 2009–2010. A recent workshop hosted by DOE recommended that if the ARM Facility were to establish additional long-term fixed sites, the highest priority should be given to placing one in the Azores due to the importance of characterizing the physical and radiative properties in these clouds and the processes that occur within them (ACRF 2007). This same report recommended that sites should be established (in relative order of priority) in Greenland to investigate the influence of Arctic clouds on the ice sheet, southeastern Asia to investigate aerosol influences on the monsoon, the Amazon rain forest to study deep tropical convection over land, and the mid-latitude southern hemisphere storm tracks to look at relatively clean clouds in an area that is difficult to simulate with numerical models. Many GCM predictions have also illustrated that the mean conditions at high latitudes are very sensitive to changes in anthropogenic emissions, and thus additional long-term data sets over the Arctic sea ice and in Antarctica would be desirable. In 2008, DOE hosted another workshop that reiterated the importance of shallow cumulus processes over the subtropical ocean and emphasized the need for further investigations in these regions (ARM 2008). These workshops have demonstrated that the need for observations in different climatic locations is great.

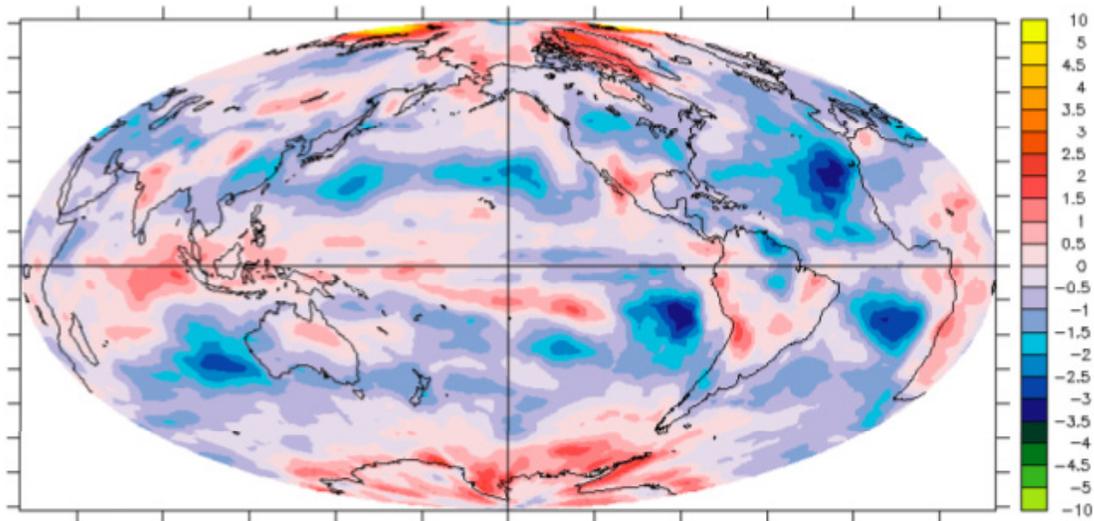


Figure 7. The regional contribution to intermodel spread in global mean cloud radiative feedback using simulations from multiple GCMs. Values close to zero indicate that there is little spread among the different model simulations, whereas larger absolute values indicate more spread. The regions that demonstrate the largest model-to-model differences in cloud radiative feedback are in the stratocumulus regions and at high latitudes. Figure from Soden et al. (2009).

The mobile facilities are extremely useful, and should be targeted to address these particular global and regional problems. However, these deployments are relatively short-term (order of 1 year), and thus the resulting data sets are best suited to sample the range of conditions that are experienced at each location rather than provide a robust data set from which climatologically representative statistics can be computed. The second mobile facility is being designed specifically for deployment in harsh marine environments, and will be used to directly tackle the large uncertainties in atmospheric processes in climate models that currently exist at many oceanic locations.

Some processes are best observed, or perhaps can only be observed, using airborne in situ techniques. The ARM Aerial Facility (AAF) is designed to make these measurements. However, previous campaigns have not fully sampled the various modes of variability that may drive the important atmospheric processes; these campaigns have typically been conducted in warm seasons during the daytime. The systematic observational capability that is being developed by the AAF should be used to more fully sample the entire diurnal, synoptic, and seasonal cycles that drive variability in the climate system. Future deployments of the AAF, like those of the mobile facilities, should be targeted directly on major problems that require the most attention, and be designed to capture these important modes of variability. Furthermore, the community needs to continue to develop new in situ instruments that can be operated from aircraft; this effort includes the miniaturization of instruments so that the instruments can be operated from smaller airframes, faster response instruments to improve spatial resolution, the hardening of laboratory instruments so they can be carried aloft, and the development of new techniques to measure geophysical variables that are currently not observed or not observed well.

In addition to new site locations and in situ capabilities, new remote sensing methods should be developed to enable better insights into many atmospheric processes. A wide range of geophysical variables needed to achieve ASR science objectives can be derived from current ARM observations,

especially considering the enormous increase in observational tools that were added to the program recently (Appendix A). However, additional important geophysical variables and processes are not well observed or perhaps can only be observed with detailed in situ observations, the latter of which do not lend themselves well to statistical analyses. Thus, it would be highly advantageous for ASR to develop new remote sensing methods to observe these elusive variables, including: profiles of aerosol absorption, profiles of CCN and IN concentrations, upper tropospheric humidity throughout the diurnal cycle in all conditions, liquid water content profiles in all clouds but especially in mixed-phase clouds, profiles of radiative and latent heating rate (currently these are only computed), four-dimensional (temporal and spatial) distributions of water vapor, and four-dimensional retrievals of aerosol properties. Developing the tools and retrievals to measure these variables is a sizeable challenge to the observational community. However, these measurements, together with those that are and can be made using the current ARM Facility instruments will further improve the ASR program's ability to characterize and understand the key processes that occur in the atmosphere along the aerosol-cloud-precipitation continuum.

Laboratory studies into the aerosol-cloud-precipitation continuum would greatly benefit from the consolidation of scattered specialized laboratories into a DOE National Atmospheric Laboratory user facility. The facility is motivated in large part by the benefit that would derive from having an aerosol and cloud chamber in which the complex set of conditions influencing clouds and aerosol are controlled. This plan has already identified a number of areas that would benefit from this facility, and in fact might require it to be fully addressed. Although ASR already supports some laboratory research (Section 3.1), it is compromised by the small size of the static chambers compared to the chambers used in several European facilities. The most prominent of these include the Leipzig Aerosol Cloud Interaction Simulator (LACIS), used for studies of aerosol-cloud interactions, the Karlsruhe Aerosol and Cloud Chamber (AIDA), and the Jülich aerosol chamber. The AIDA chamber uses an expansion cooling that enables it to be a center of excellence for ice nucleation and ice crystal growth investigations. The Jülich chamber is being used to investigate new particle formation involving isoprene—a hydrocarbon that dominates global emissions from vegetation—and for placing these studies in the context of global warming (Ziemann 2009). U.S. scientists have had to simulate these processes under less realistic conditions. What is clearly needed to advance understanding of cloud as well as aerosol formation is a dedicated consolidated laboratory facility in the U.S. capable of conducting experiments under controlled conditions. In addition to the aforementioned chamber-type studies, one can envisage including wind tunnels for designing probe inlets for in situ aircraft sampling of aerosol and hydrometeors (e.g., to address the problems associated with ice shattering at inlets), for ice fall velocity measurements, and for measuring the aerodynamic breakup of droplets and natural ice multiplication mechanisms. Such a facility would be a bold, breakthrough-science-type initiative that would lay a firm foundation for systematically improving cloud and aerosol process/properties modules as well as serve as an incubator for the development of new cloud and aerosol instruments designed for field deployment.

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Appendix A ARM Instrumentation

The ARM Climate Research Facility has a wide arrange of in situ and remote sensing tools that are able to provide a detailed characterization of the cloud, aerosol, precipitation, dynamic, thermodynamic, and radiative properties of the atmosphere. The ARM Facility was greatly enhanced by the American Recovery and Reinvestment Act (ARRA), which allowed some critical but aging instruments to be replaced with more modern versions and providing new capability to observe properties and processes that were unable to be observed before.

The following table provides one view of the ARM instrumentation that will be used by the ASR program. The new capability provided by ARRA is indicated in red; note that in some cases a new instrument was added to the entire facility, whereas in others an instrument already existed at some ARM sites and was added to other sites that did not have that instrument. The table indicates the primary variables that the instruments measure and the important geophysical variables that can be derived or retrieved from these measurements. Note that this table does not include a list of geophysical variables that can be derived using a combination of data from multiple instruments.

The various ARM sites are: SGP – Southern Great Plains; TWP – Tropical Western Pacific; NSA – North Slope of Alaska; AMF – ARM Mobile Facility (refers to both here); AAF – ARM Aerial Facility; and MAOS – Mobile Aerosol Observing System. Instruments marked with a dagger (†) are replacements/upgrades of aging systems that were nearing the end of their lifetimes.

Instrument	Location	Primary Variables Measured	Primary Geophysical Variables Derived
Millimeter-wave cloud radar	SGP, NSA, TWP, AMF	8.6 mm wavelength radar reflectivity, Doppler velocity, Doppler spectral width	Cloud boundaries, cloud liquid and ice water content profiles, cloud particle motion
Millimeter-wave cloud radar	AMF	3.0 mm wavelength radar reflectivity, Doppler velocity, Doppler spectral width	Cloud boundaries, cloud liquid and ice water content profiles, cloud particle motion
Scanning dual-frequency cloud radar	SGP, NSA, AMF	8.6 mm and 3.0 mm wavelength radar reflectivity, Doppler velocity, Doppler spectral width, scans in both elevation and azimuth	Cloud boundaries, cloud liquid and ice water content profiles, cloud particle motion, cloud liquid water content profiles
Scanning dual-frequency cloud radar	TWP, AMF	3 cm and 8.6 mm wavelength radar reflectivity, Doppler velocity, Doppler spectral width, scans in both elevation and azimuth	Cloud boundaries, cloud liquid and ice water content profiles, cloud particle motion, cloud liquid water content profiles
Scanning precipitation radar	SGP, NSA, TWP, AMF	Different wavelengths for different sites (3 to 8 cm), profiles of radar reflectivity, Doppler velocity, and Doppler spectral width, scans in elevation and azimuth	Rain rate, cloud motion

Instrument	Location	Primary Variables Measured	Primary Geophysical Variables Derived
Radar wind profilers	SGP, NSA, TWP, AMF, MAOS	UHF wavelength (~1000 MHz) signal to noise ratio profiles, wind speed and direction profiles	Boundary layer depth
Radiosonde	SGP, NSA, TWP, AMF	Profiles of temperature, humidity, wind direction, wind speed, and pressure	Precipitable water vapor
Raman Lidar	SGP, TWP	Water vapor mixing ratio profiles, aerosol and cloud scattering and extinction profiles, depolarization profiles	Relative humidity profiles, cloud base height
Sonic detection and ranging system	MAOS	Wind profiles	
Doppler wind lidar	SGP, TWP, AMF	Profiles of vertical velocity	Boundary layer turbulence, updraft velocity
High-spectral-resolution lidar	NSA, AMF	Profiles of aerosol and cloud backscatter and extinction, depolarization ratio	Cloud boundaries, cloud optical depth, cloud phase
Micropulse lidar	SGP, NSA, TWP, AMF†	Lidar backscatter profiles at 532 nm, depolarization profiles	Cloud boundaries, cloud optical depth, cloud phase
Ceilometer	SGP, NSA, TWP, AMF†	Cloud base height below 7 km	
Microwave radiometer	SGP, NSA, TWP, AMF†	Downwelling microwave brightness temperature at 23.8, 31.4, and 90.0 GHz	Precipitable water vapor, cloud liquid water path
Low water vapor microwave radiometer	NSA	Downwelling microwave brightness temperature at ±1, 3, 7, 12 GHz from the 183.3 GHz water vapor line	Precipitable water vapor in very dry conditions, liquid water path
Microwave radiometer profiler	SGP, AMF	Downwelling microwave brightness temperature at 12 frequencies between 22.2 and 58 GHz	Precipitable water vapor, liquid water path, low-vertical-resolution profiles of water vapor and temperature
Low water vapor microwave profiler	NSA	Downwelling microwave brightness temperature at 15 frequencies between 170 and 183.3 GHz	Precipitable water vapor in very dry conditions, liquid water path, low-vertical-resolution profiles of water vapor in dry conditions
Atmospheric emitted radiance interferometer	SGP, NSA, TWP, AMF†	Downwelling infrared radiance from 3-19 μm at high spectral resolution	Temperature and humidity profiles, cloud optical depth, cloud particle effective radius
Infrared thermometer	SGP, NSA, TWP, AMF	Downwelling infrared radiance at 10 μm, upwelling infrared radiance at 10 μm	Surface skin temperature

Instrument	Location	Primary Variables Measured	Primary Geophysical Variables Derived
Total sky imager	SGP, NSA, TWP, AMF	Hemispheric visible sky images	Cloud fraction
Longwave broadband radiometers	SGP, NSA, TWP, AMF	Downwelling and upwelling hemispheric infrared flux (4-50 μm)	Infrared cloud forcing and effective cloud fraction
Shortwave broadband radiometers	SGP, NSA, TWP, AMF	Downwelling hemispheric diffuse and direct-beam visible/near-infrared flux (0.3 - 4 μm), upwelling hemispheric visible/near-infrared flux	Visible/near-infrared cloud forcing and effective cloud fraction
Normal-incidence multi-filter radiometer	SGP	Direct beam irradiance in 6 10-nm wide channels from 440 nm to 940 nm	Aerosol optical depth, aerosol Angstrom coefficient, precipitable water vapor
Multi-filter rotating shadowband radiometer	SGP, NSA, TWP, AMF	Downwelling hemispheric diffuse and total irradiance in 6 10-nm wide channels from 440 nm to 940 nm	Aerosol optical depth, aerosol Angstrom coefficient, precipitable water vapor
Shortwave spectrometer	SGP	Downwelling visible/near-infrared radiance from 300 to 1800 nm at 4-8 nm spectral resolution	Aerosol and cloud optical depth
Solar spectrometer	SGP, AMF2	Solar spectral radiance and irradiance	Aerosol and cloud optical depth
Narrow field-of-view radiometer	AMF	Downwelling zenith radiance at 660 and 870 nm	Cloud optical depth, effective cloud fraction
Cimel sunphotometer	SGP, NSA, TWP, AMF†	Downwelling sky radiance and solar irradiance in 6 narrow spectral channels from 340 nm to 1020 nm	Aerosol optical depth, aerosol Angstrom coefficient, aerosol size distribution, aerosol single scatter albedo
Aerosol observing system	SGP, NSA, TWP, AMF, MAOS, AAF†	Surface-based in situ measurements of aerosol scattering and backscattering as a function of relative humidity, absorption, number concentration, size distribution, and hygroscopic growth, tandem differential mobility analyzer, multi-supersaturation cloud condensation nuclei counter	Aerosol single scatter albedo, aerosol asymmetry parameter, hygroscopic growth
Particle mass spectrometers	SGP, TWP, MAOS, AAF	Inorganic and volatile organic aerosol chemical composition	
Aircraft Integrated Meteorological Measurement System	AAF	5-port air motion sensing: true air speed, altitude, angle-of-attack, side-slip - and temperature, relative humidity	

Instrument	Location	Primary Variables Measured	Primary Geophysical Variables Derived
Particle into liquid sampler	MAOS, AAF	Aerosol chemical composition, water soluble organic carbon	
Scanning mobility particle sizer	MAOS, AAF	Aerosol size distribution from 15 nm to 450 nm	
Cloud in situ probes	AAF	Cloud droplet size distribution, cloud particle images, cloud water content, cloud particle phase	
Proton transfer reaction mass spectrometer	MAOS	Gaseous organic compounds	
Ozone	MAOS, AAF, TWP	Ozone concentration	
Trace gas precursors	MAOS, AAF	Carbon monoxide, sulfur dioxide, nitrous oxide, nitrogen dioxide	
Meteorological observations	SGP, NSA, TWP, AMF, MAOS†	Surface- and tower-based observations of pressure, temperature, relative humidity, wind speed, wind direction, and precipitation amount	
2-Dimensional Video Disdrometer (2DVD)	TWP, AMF, MAOS	Drop size distributions, precipitation	Radar reflectivity, liquid water content
Disdrometer	SGP, TWP†	Surface-based rain droplet size distribution, rain rate	Radar reflectivity, liquid water content
Optical rain gauge	SGP	Surface-base rain rate and accumulation	
Eddy correlation flux measurement systems	SGP, AMF, TWP, NSA†	Surface latent heat flux, surface sensible heat flux, atmospheric turbulence, carbon dioxide flux	
Energy balance Bowen ratio stations	SGP	Surface latent heat flux, surface sensible heat flux	Bulk aerodynamic fluxes
Soil water and temperature system	SGP	Soil moisture and temperature measurements at 6 depths from 5 cm to 125 cm	

Appendix B

Accomplishments of the ARM and ASP Programs

The goal of this appendix is to provide one view of the accomplishments of the DOE observation-based climate research programs. These contributions to climate change research would not be possible without the high quality, long-term data provided by the ARM Climate Research Facility and the laboratory and periodic field studies performed by the ASP Program. The ASR website (at <http://asr.science.energy.gov>) will provide a comprehensive list of research performed under the auspices of the ARM and ASP programs, including a publications database that lists nearly 2000 peer-reviewed journal articles whose authors have made use of the ARM and ASP data for climate research. While the accomplishments of the two programs are too numerous to mention here, this appendix summarizes several of the more important contributions.

Establishing a new standard for climate research observations. ARM was the first climate research program to deploy a suite of cutting-edge instrumentation for obtaining continuous measurements of cloud and aerosol properties (Ackerman and Stokes 2003). This strategy revolutionized our ability to collect long-term statistics of detailed cloud properties and now serves as a model for programs around the world (Clothiaux et al. 2000; Kollias et al. 2007; Illingworth et al. 2007). The ARM Facility paradigm of long-term continuous measurements is essential to the enhancement and evaluation of climate models that must simulate the evolution of atmospheric properties for long continuous periods, from decades to centuries. This measurement approach permits unparalleled examination of atmospheric process behavior and model performance evaluation over extended periods and a wide range of meteorological conditions.

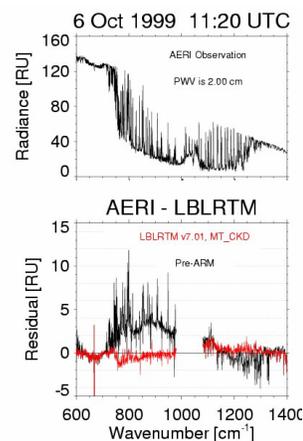
Obtaining aerial measurements to supplement ground-based observations. Observations at fixed and mobile sites are supplemented periodically with observations from aircraft. These data have yielded insights into a range of science issues including the absorption of radiation by clouds (e.g., Valero et al., 2003), aerosol properties (e.g., Andrews et al. 2004; Berg et al. 2009), and cloud properties (e.g., Um et al. 2009). Detailed cloud properties such as ice crystal sizes are critical because they dictate the life cycle of a cloud and its interaction with radiation. Airborne measurements obtained during ARM Facility field campaigns in the Arctic (McFarquhar and Cober 2004), the tropics (May et al. 2008), and mid-latitudes (Mace et al. 2002) revealed new information about ice crystal sizes and shapes in various cloud types. Several aircraft campaigns have focused on the life cycle of aerosol particles in the atmosphere, in order to better understand the evolution of the chemical and microphysical properties (e.g., Molina et al. 2008; Ferrare et al. 2006). Recently, the program has begun long-term airborne (multi-month) campaigns; these routine missions allow for improved gathering of statistics and sampling of the range of natural conditions. These observations led to greatly improved techniques for retrieving cloud and aerosol properties from the ground, as well as providing insight into the processes occurring within the atmosphere.



A Twin Otter flying taking off to fly a routine mission over the SGP during a long-term experiment in 2009.

Achieving significant improvement in water vapor measurements. While carbon dioxide is a key contributor to climate change, the dominant greenhouse gas in the Earth’s atmosphere is water vapor. Using detailed ARM Facility measurements of water vapor and associated radiative transfer calculations, ASR scientists have reduced uncertainty in water vapor measurements from 13% to less than 4% during the past decade (Revercomb et al. 2003, Turner et al. 2003). Large improvements in the measurement of upper tropospheric humidity, both by radiosondes and using remote sensors such as Raman lidar, have also been realized over the last decade (Ferrare et al. 2004, Soden et al. 2004); these improved observations allow the investigation of the frequency of supersaturation in the upper troposphere and the connection with cirrus nucleation (Comstock et al. 2004).

Improved detailed models of radiation transfer. Spectrally resolved ARM Facility radiation measurements and improved water vapor observations have been central to improvements in the parameterization of the amount of longwave radiation absorbed by the water vapor continuum (Clough et al. 1989). These improvements, generally from small incremental changes, were made primarily in the water vapor self- and foreign-broadened continuum and the water vapor absorption line parameters. These changes, when taken as a whole, result in up to a 6 W/m² improvement in the modeled clear-sky downwelling longwave radiative flux at the surface, as well as significantly better agreement with spectral observations, resulting in the agreement between theory and observation of better than 1% (Turner et al. 2004). Similar advancements have been made to the detailed shortwave radiative transfer models, with the observations demonstrating that there are no significant unknown gaseous absorbers in the shortwave or longwave portions of the spectrum (Mlawer et al. 1998, Ackerman et al. 2003). Current shortwave radiative transfer models can now compute the clear sky downwelling shortwave flux with an accuracy of better than 2% (Michalsky et al. 2006). The ASR program also developed a method that has been proposed to the World Meteorological Organization as a standard for diffuse horizontal shortwave irradiance (Michalsky et al. 2007).



The downwelling infrared radiance at the SGP site, together with observed minus computed radiance residuals for two cases: pre-ARM (circa 1990) in black, and circa 2003 in red.

Radiative heating in the upper troposphere. Infrared radiation is associated with wavelengths from approximately 4 to 100 μm . The accuracy of infrared radiative transfer models in the 4-15 μm region is better than 1% (Turner et al. 2004); however, the far-infrared (wavelengths longer than 15 μm) is very uncertain, due to lack of spectrally resolved measurements in this band and strong water vapor absorption making the spectral region opaque at most locations. The radiative heating of the middle and upper troposphere is dominated by emission from the far-infrared, and inaccuracies in radiative transfer models in this spectral region affect the vertical motion of the atmosphere (and hence the general circulation) as well as the total outgoing longwave radiation of the planet. To collect the data needed to evaluate and improve radiative transfer models in the far-infrared, scientists conducted the Radiative Heating in Underexplored Bands Campaigns (RHUBC) (Turner and Mlawer, 2009). Recently developed spectrally resolved far-infrared radiance instruments and very sensitive water vapor radiometers were deployed at the ARM Facility site in Barrow and at a high-altitude site (above 5 km mean sea level). While the analysis of these data sets is still ongoing, there have already been important modifications to spectral

absorption models that translate directly into better water vapor retrievals and improved ability to simulate the radiative transfer in the far-infrared (Payne et al. 2008, Delamere et al. 2009).

Radiative heating of the atmosphere by clouds. Clouds can have a major effect on the radiative heating of the atmosphere. To understand the influence of clouds, scientists must understand the cloud radiative heating effect on the general circulation of the atmosphere. With the retrieval of cloud properties on a continuous basis and the improvement of radiative transfer models, ASR scientists have derived climatologies of the vertical profiles of radiative heating at the ARM Facility sites. These heating rate profiles are essential for testing radiation and cloud parameterizations in GCMs, as they provide a strong constraint on the vertical distribution of heating within the atmospheric column. Work in this area has provided a remarkably detailed data set for studying the redistribution of radiative energy in the atmosphere (Mace et al. 2006; Mace and Benson 2008; Mather et al. 2007), making it possible to evaluate cloud and radiative profiles in climate models (McFarlane et al. 2007), and allowing the determination of the characteristic radiative heating profile associated with different cloud types (Mather and McFarlane 2009).

Implementing a major improvement in the radiative effects of clouds in GCMs. Climate models have a particularly difficult time representing complex cloud systems in a relatively coarsely resolved spatial domain. Examination of cloud fields observed at the ARM Facility sites, combined with efforts to represent the radiative effects of those cloud fields, led to the development of a new cloud radiation scheme adopted for use in several climate models and weather forecasting models (Pincus et al. 2003, 2006; Räisänen et al. 2005, 2007; Morcrette et al. 2008). This approach uses stochastic methods to account for the clouds in a partially cloudy atmosphere, and the results demonstrate much better agreement with the observed fluxes than previous methods.

Improving the representation of radiation in climate models. ASR scientists used ARM Facility observations to significantly improve calculations of atmospheric distribution of radiant energy. These improvements are encapsulated in the Rapid Radiative Transfer Model (Mlawer et al. 1997). Because this model offers greater accuracy and efficiency, it has been incorporated into several climate and numerical weather prediction models (Iacono et al. 2000; Morcrette et al. 2008). Advancements in radiation calculations in these global models have led to improved forecasts of temperature and humidity in the upper atmosphere (Iacono et al. 2003).

Providing unique observations of the radiative impact of aerosols. While the effect of greenhouse gases is well characterized in GCMs, uncertainty remains regarding the effect of aerosols, such as dust and smoke. This is particularly true regarding the effect aerosols have on radiation transfer in the atmosphere, either by redirecting incoming solar radiation back into space or by redirecting outgoing infrared radiation toward the surface. Extensive aerosol observations from the ARM Facility sites and ARM Mobile Facility have quantified the impact of aerosols on the radiation budget in diverse climatic regions (e.g., Ferrare et al. 2006). ARM made the first column radiation absorption measurements of the impacts of Saharan dust (Slingo et al. 2006).

Atmospheric radiative balance in the subtropics. In 2006, the AMF was deployed in Niamey, Niger for the ARM campaign Radiative Divergence Using AMF, GERB, and AMMA Stations, or RADAGAST (Miller and Slingo, 2007). This campaign was part of a large, multi-year international field program, titled the African Monsoon Multidisciplinary Analyses (AMMA) Project. RADAGAST was designed to supply continuous measurements of the broadband upward and downward solar and thermal radiative fluxes at the surface. When combined with corresponding measurements from the Geostationary Earth Radiation Budget (GERB) instrument on a European satellite, these measurements provide the first well sampled direct estimates of the energy balance across the atmosphere. RADAGAST played a major role in linking multi-scale observations, data analysis, and modeling, allowing both weather and climate models to better predict the formation and dynamics of the West African monsoon.



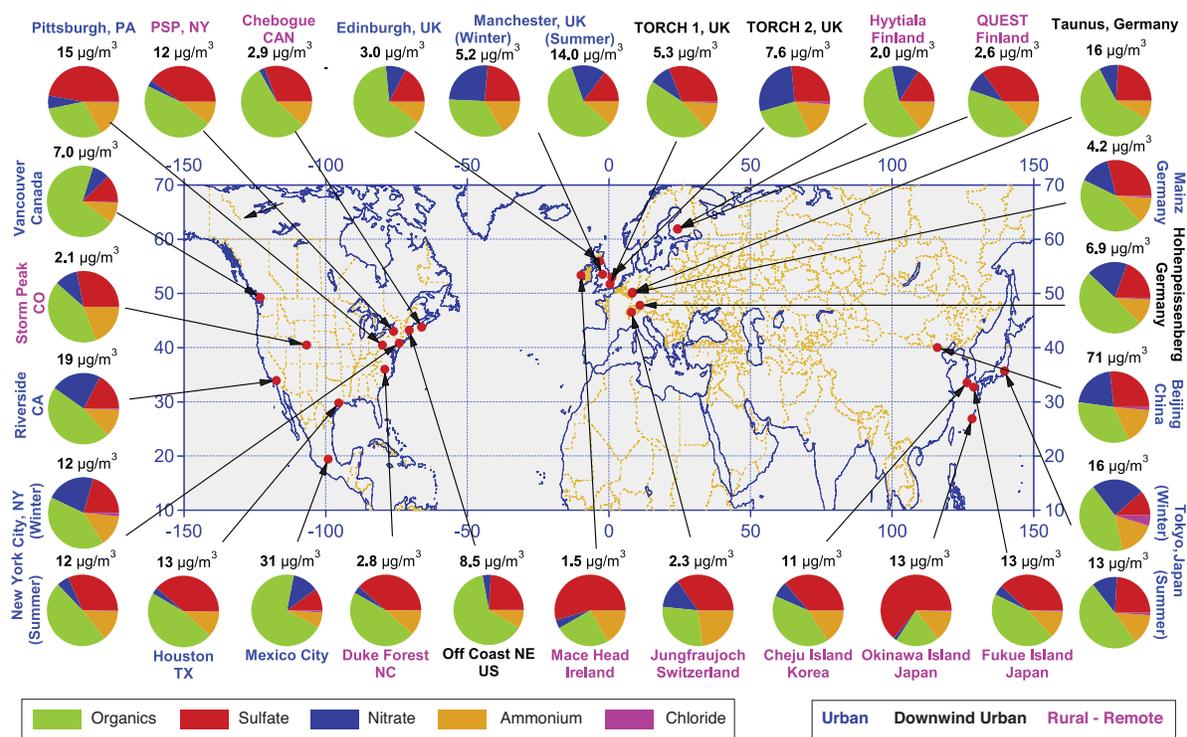
In March 2006, the ARM Mobile Facility and satellite instruments made the first simultaneous observations of a major dust storm from space and the ground, allowing researchers to test their understanding of how dust affects the radiant energy budget of the atmospheric column.

The RADAGAST experiment provided a unique data set to characterize the thermodynamic, radiative, and aerosol characteristics of the Niamey region (Slingo et al. 2008, 2009). A huge dust storm was observed during the campaign with a dust optical depth larger than 3, resulting in solar heating of the atmosphere of over 400 W/m² (Slingo et al. 2006). RADAGAST data demonstrated that dry season aerosols exhibited different characteristics when compared to wet season aerosols, and that the characteristics of a dust-laden aerosol are quite different from a smoke-laden aerosol (Turner 2008; McFarlane et al. 2009), both of which had significant impacts on the seasonal energy balance at the surface (Miller et al. 2009a).

The sensitivity of direct aerosol radiative forcing. Aerosol direct radiative forcing depends on aerosol optical properties (optical depth, single scattering albedo, and asymmetry parameter) and on the surface albedo in ultraviolet-visible and shortwave infrared spectral ranges. Uncertainties in measurements in these input parameters produce uncertainty in estimates of the aerosol direct radiative forcing. To quantify this uncertainty, McComiskey et al. (2008) calculated the aerosol radiative forcing at the top of the atmosphere and surface for conditions typical at the three ARM Facility permanent sites: Tropical Western Pacific (TWP), Southern Great Plains (SGP), and North Slope of Alaska (NSA), using values of the parameters typical for each site. For measurement uncertainties used in the study, uncertainty in total modeled forcing at the top of the atmosphere ranges from approximately 0.6 to 1.1 W m⁻².

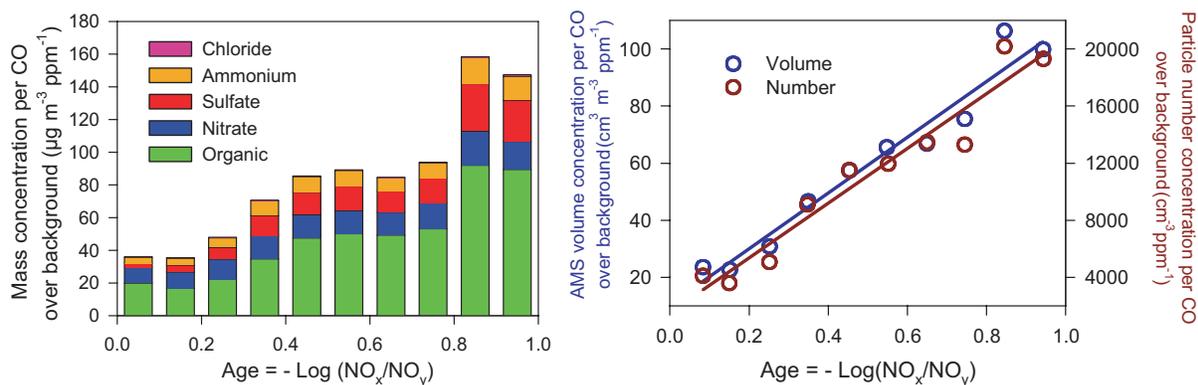
Ubiquity and dominance of oxygenated species in organic aerosols. A new aerosol mass spectrometer (developed in part with support from ASP and the DOE SBIR program) that permits examination of the organic and inorganic mass concentration and composition of size-selected or total aerosol has been used in multiple field studies over much of the Northern Hemisphere. These measurements have led to two generalizations about aerosol composition that greatly extend knowledge of aerosol composition and understanding of the processes controlling this composition. First, measurements with the aerosol mass spectrometer by many investigators show that organics are the major or dominant aerosol constituent at

urban, downwind-urban, and rural locations throughout the anthropogenically influenced Northern Hemisphere. Second, the organic fraction of the aerosol increases with increasing distance from urban sources, suggesting anthropogenic enhancement of the organic material. Examination of relative abundances of specific mass fragments that can be associated with primary versus secondary aerosol substances demonstrates the increase in secondary material with distance from urban emission sources (Zhang et al. 2007).



Aerosol composition measured at the surface at various locations around the world (Zhang et al. 2007).

Secondary organic aerosol production in a mega-city plume. ASR research has demonstrated major limitations in current understanding of secondary organic aerosol (SOA) formation. Aircraft measurements of the amount and composition of aerosol during the 2006 MAX-MEX field campaign in Mexico City, normalized to mixing ratio of carbon monoxide (a conservative tracer of urban emissions) to account for dilution, were stratified according to photochemical age, in order to examine atmospheric evolution (Kleinman et al., 2007, 2008) yielding the time development of aerosol properties. SOA formation causes a 5-fold increase in organic aerosol in less than one day. The measured increase in organic aerosol exceeds the modeled increase (based on measured volatile organic carbon) 10-fold. The increase is nearly identical to that observed in the eastern U.S., despite the expectation that SOA yields should increase more in areas with higher concentrations of aerosol precursors.



Left: Mass concentrations of aerosol species, normalized to excess CO, from aircraft measurements in the Mexico City Plume throughout the 2006 MAX-MEX field campaign as function of photochemical age. Right: similar plot for aerosol volume and accumulation mode number concentration with linear regression lines. Volume and number scales are proportional, both indicating a ~ 5 -fold increase with age over period corresponding to approximately one day atmospheric residence time.

New processes for formation and loss of secondary organic aerosol. As noted in the paragraphs above, an important issue driving scientific uncertainty in aerosol forcing is the observed production of secondary organic aerosol mass in substantially greater quantities than can be explained by current models. Several ASP laboratory studies were undertaken to address this discrepancy; some of the major findings are described in the following paragraphs.

Because of its low molecular weight, the biogenic precursor isoprene has not previously been considered a major source of SOA; however, recent laboratory studies by ASP investigators have shown a small yield of SOA from isoprene oxidation. Because of isoprene's large source strength in deciduous forests, even a small yield of SOA could constitute a major source of SOA. Examination of SOA from isoprene in a global chemical transport model showed substantial enhancement of SOA in forested regions at the surface and much greater relative enhancement in the free troposphere (Henze and Seinfeld 2006). More recent work (Paulot et al., 2009) shows a previously unrecognized pathway from isoprene emissions to SOA formation. Global simulations show an enormous flux—nearly 100 teragrams of carbon per year—to the atmosphere, providing a missing link tying the gas-phase degradation of isoprene to the observed formation of organic aerosols. This work suggests a potential resolution of the large discrepancy between measurement and model-based estimates of SOA formation and also suggests a possible role of these organic aerosols as cloud condensation nuclei.

Incorporating both direct and indirect effects of particles on climate requires a full understanding of the processes that lead to SOA formation as well as possible loss processes. ASR researchers discovered a new process for formation of SOA that involves the oxidation of organics due to photochemistry in and at the surface of particles. Their research showed that α -pinene, a major SOA precursor through its reactions with O_3 and to a lesser extent OH and NO_3 , was oxidized at the air-water interface of nitrate solutions during photolysis (Yu et al. 2008 a,b). This process led to the formation of a number of products in the particles, including pinic and pinonic acids and trans-sobrerol.

This new mechanism of SOA formation may be related to the observation by Smith et al. (2008) that demonstrated that nitrate and organics were both significant components of the smallest particles yet to be analyzed in air (sizes as small as 6 nm). Recent theoretical studies (Miller et al. 2009b) show that nitrate ions in small clusters, up to ~300 water molecules, prefer to reside at the air-water interface where this photochemistry will be enhanced. These small clusters approach the size of new particles as they form, suggesting that this newly discovered mechanism for SOA formation may be important during nucleation and initial particle growth.

A laboratory study (Song et al. 2007) of SOA formation from oxidation of α -pinene (a major biogenic SOA precursor gas) revealed a major discrepancy with a semi-empirical approach commonly used to represent SOA formation in global aerosol models. The presence of primary organic aerosol (POA) seed particles was found not to enhance the SOA yields. However, in contrast to the mass yields, the seed particles had a great effect on the aerosol dynamics, as reflected in the particle size distributions. If these results are found to apply to other biogenic SOA precursors, then anthropogenic POA emissions would have a much lower direct impact on the global biogenic SOA mass budget than previously thought. However, they could potentially decrease the number concentrations of purely biogenic SOA particles in forested regions impacted by urban sources.

Model for Simulating Aerosol Interactions and Chemistry. An ongoing priority of ASR is to improve and implement model descriptions of major atmospheric processes. In this regard, atmospheric aerosol chemistry is especially challenging. The coupled ordinary differential equations for dynamic gas-particle mass transfer are extremely “stiff.” Consequently, accurate numerical models of aerosol mass transfer are computationally very expensive to run. ASR Scientists have overcome these limitations with a new aerosol model called MOSAIC (Zaveri et al. 2008). It uses a mass-transfer solver called “Adaptive Step Time-split Euler Method,” or ASTEM. Several performance tests showed MOSAIC to exhibit excellent agreement with a benchmark version of the model that used a rigorous solver for integrating the stiff equations. In addition, MOSAIC/ASTEM was found to be ~100 times faster than the rigorous solver, and the steady-state MOSAIC results for monodisperse aerosol test cases were found to be in excellent agreement with those obtained with the benchmark equilibrium model, the Aerosol Inorganics Model. These results showed that MOSAIC is extremely efficient without compromising accuracy, and therefore is highly attractive for use in regional/global aerosol models (e.g., Chapman et al. 2008).

Interactions of Aerosols with water vapor and cloud processes. The spectrum of the effects that aerosol indirect effects have on climate is thought to be of greater magnitude and certainly of greater uncertainty than aerosol direct forcing effects. Accordingly, the interaction of atmospheric aerosols with water vapor and clouds is a major focus of effort for ASR. The following paragraphs highlight several program accomplishments in this important area.

Impact of organic coatings on hygroscopic salt particles. In ASR laboratory studies, the presence of even a thin, sub-monolayer amount of organics on deliquesced NaNO_3 particles was shown to significantly increase water retention (Zelenyuk et al., 2009). This resulted in smaller measured particle densities than expected for NaNO_3 , even at amounts of organics too small to be seen by single particle mass spectrometry. This dramatic effect of an undetectable sub-monolayer amount of organics on particles has important implications for interpretation of the measurements of density and composition of airborne particles. In addition, water retention and formation of organic shells can potentially impact light scattering by these particles, as well as their ability to act as cloud condensation nuclei (CCN).

Formation of CCN from secondary organic aerosol. Smith et al. (2008) measured the composition, hygroscopicity, and CCN activation of small aerosol particles above Mexico City. New particle formation, identified by a great increase in concentration of particles below 10 nm diameter, was attributed to photochemical activity triggered by ultraviolet radiation with a composition dominated by organics. Prior to the new particle formation event, measurements of hygroscopic growth showed that particles were quite hydrophobic, increasing in diameter by a factor of only 1.1 as relative humidity was increased to 90%. Few of the 40 nm particles could activate into cloud droplets even at supersaturations as high as 1%. During the event, however, the growth factor jumped to 1.7, typical of highly hygroscopic substances such as ammonium sulfate, and 40 nm particles could activate at 0.5% supersaturation. This result shows that the freshly nucleated particles can rapidly grow into the size range that is effective as CCN and, despite being comprised mostly of organics, can be highly hygroscopic and highly efficient as CCN, challenging conventional thinking that CCN are predominantly inorganic and that organics suppress cloud droplet formation.

Closure study on CCN concentration. Most current physically based attempts to represent aerosol effects on warm clouds are based upon Kohler theory, which expresses the critical supersaturation for activation of an aerosol particle as a function of its diameter and hygroscopicity. To test Kohler theory, Wang et al. (2008) used data taken on nine aircraft flights during the ASP Marine Stratus Experiment off the coast of northern California. The accuracy of Kohler theory was examined by comparing calculated and measured number concentrations of cloud condensation nuclei CCN. For aerosol with organics volume fraction up to 70%, such as the marine boundary layer and free troposphere aerosols, CCN concentration was found to be insensitive to the hygroscopicity of the organics and can be accurately calculated with a constant hygroscopicity for all organic species. However, for the aerosol within thin layers above clouds, for which organics contributed up to 90% of the total aerosol volume, an accurate knowledge of the overall organic hygroscopicity was required to accurately calculate CCN concentrations.

The first ground-based remotely sensed evidence of indirect aerosol impacts on clouds. The first aerosol indirect effect of suspended atmospheric particles on the microphysics of clouds occurs when an increase in the number of aerosol or CCN within a cloud causes a decrease in the average cloud droplet size, reflecting more solar energy to space. The first ground-based remote sensing measurements of this phenomenon were made at the SGP site (Feingold et al. 2003). A more detailed look at this indirect effect was performed during the May 2003 Aerosol IOP, a field campaign conducted at the SGP site, in which both in situ and remote sensing instruments were available to measure aerosol and cloud parameters. The measurements confirmed the presence of the first aerosol indirect effect, but also showed that the methodology used for retrieving droplet size and the proxy used for aerosol, or CCN, concentration strongly affected the magnitude of the drop size response to changes in aerosol (Feingold et al. 2006). These results have guided additional studies that have used longer-term ground-based data sets to quantify the aerosol indirect effect (Kim et al. 2008; McComiskey et al. 2009). Additionally, data from the Aerosol IOP were used to predict CCN concentration at cloud altitude from ground-based in situ remote sensing observations (Ghan et al. 2006).

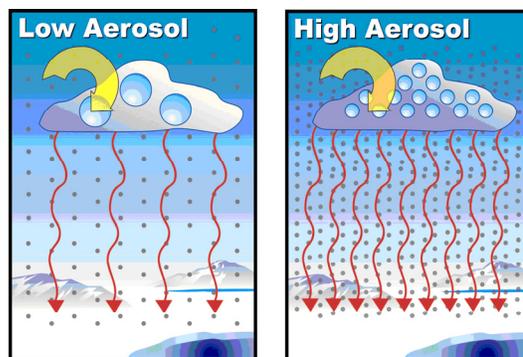
Quantifying the radiative impact of aerosols on small liquid water clouds. Thin clouds with a liquid water path (LWP) below 50 g/m² cover approximately 30% of the globe, playing an important role in the Earth's radiation budget. Radiative fluxes at the Earth's surface and top of atmosphere (TOA) are very sensitive to LWP variation when the LWP becomes smaller than ~50 g/m² (Turner et al. 2007). Thus,

aerosol effects on thin clouds can substantially impact the variation of global radiative forcing if LWP changes. In cases of thin warm stratocumulus clouds, increased aerosols lead to increased cloud droplet number concentration, providing increased surface area of droplets where water vapor condenses. This increases condensation, and thus condensational heating, producing stronger updrafts and leading to an increased LWP with increased aerosols in cases where precipitation reaches the surface (Lee et al. 2009). The resulting change in the size of the cloud droplets also has a significant impact on the radiative flux at the cloud level, as well as at the surface and top of the atmosphere. In a case with no surface precipitation, LWP decreases with increases in aerosols. In this case, most precipitation evaporates just below the cloud base. With decreasing aerosols, precipitation increases and leads to increasing evaporation, thereby increasing instability around cloud base. However, there are many other factors besides aerosol concentration that influence cloud development, such as cloud-scale vertical motions and entrainment. Therefore, it is challenging to develop robust statistics on the first aerosol indirect effect on clouds (Lu et al. 2007).

Identification of an aerosol indirect effect in the longwave. At the NSA site, ASR scientists used ARM Facility observations to demonstrate that enhanced aerosol concentrations cause cloud droplets to be smaller and more numerous within clouds of fixed water amount, thereby affecting the longwave radiative fluxes (most studies of the aerosol indirect effect focus solely on the shortwave radiative impact). A 6-year analysis at the NSA site found that this process can make clouds more opaque, causing them to emit more thermal energy to the surface (Lubin and Vogelmann 2006). This insight is significant for understanding the Arctic energy balance.

Arctic cloud-aerosol interactions. Clouds play a particularly important role in the surface energy balance in the Arctic and are difficult to model. Several recent studies suggest that the Arctic climate is more sensitive to changes in climate forcing than other regions on Earth, while global climate models are less reliable in this region (ACIA 2004). Data from the highly successful Mixed-Phase Arctic Cloud Experiment (M-PACE, Verlinde et al. 2007) documented the microphysical structure of Arctic mixed-phase clouds, which typically consist of a stratiform layer of supercooled liquid water from which ice crystals form and precipitate (e.g., McFarquhar et al. 2007). Cloud water and ice were observed to covary directly with in-cloud vertical velocities, suggesting important microphysical linkages between the formation of cloud ice and liquid water in these clouds (Shupe et al. 2008).

M-PACE measurements also demonstrated that the number of ice nuclei in the region is much lower than assumed in standard climate models, which impacts the ability of these models to properly simulate the clouds and thus radiant energy fluxes (Prenni et al. 2006). However, even when accounting for a high degree of experimental uncertainty, the number of ice crystals measured during M-PACE could not be explained with measured ice nuclei and known ice formation mechanisms, suggesting that some ice nuclei may not be detectable by current instruments and/or that large-scale numerical weather prediction and global climate models may lack representation of some dominant ice formation mechanisms (Fridlind et al. 2007, Morrison et al. 2008, Fan et al. 2009). An international model intercomparison study based



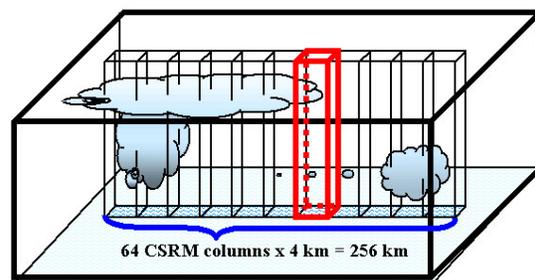
The larger aerosol concentration results in smaller cloud droplets, which increase the emission of the cloud towards the surface when the cloud is semi-transparent in the infrared.

on M-PACE data, which attracted the largest number of participants in such a study to date, showed that predictions from all types of models differed widely from one another and from the observations under common Arctic conditions (Klein et al., 2009; Morrison et al., 2009). Key differences among model sensitivities to ice nucleus concentration were identified as attributable to ice habit and fall speed specifications, underlining the importance of further developing detailed ice property measurements using ground-based remote-sensing instruments to guide model improvements (Avramov and Harrington 2009). A follow-up experiment called the Indirect and Semi-direct Aerosol Campaign (ISDAC) was conducted in the spring of 2007, providing observations of polluted springtime Arctic conditions to contrast with the seasonally clean autumnal Arctic conditions observed during M-PACE.

Developing a new paradigm for using observations to improve climate models. The detailed and comprehensive measurements obtained at the ARM Facility sites are critical for model evaluation and improvement. ASR scientists developed a unique process to bridge the gap between observations and GCMs. In this process, a subset of ARM Facility observations are combined (Zhang et al. 2001) to provide input to a GCM, while other ARM Facility observations, such as cloud profiles, are used to evaluate the model’s performance (Randall et al. 1996). This technique has led to specific improvements in GCMs, including the treatment of ice crystals in cirrus clouds (Liu et al. 2007).

Translating detailed atmosphere observations for climate modelers. Many ARM Facility instruments generate information that requires specialized skills to understand and apply. ARM Facility staff and collaborators in the research community are working together to take this complex information and generate simple physical parameters that are readily accessible by the climate modeling community. One recently developed product is the Cloud Modeling Best Estimate (CMBE), which combines a set of cloud observations from various ARM Facility instruments on a common grid (Xie et al. 2009). This product is expected to greatly facilitate the use of ARM Facility cloud data by climate modelers and has been adopted as a standard evaluation tool by the National Center for Atmospheric Research Community Atmosphere Model (CAM).

Developing a revolutionary new approach to climate modeling. Typically, GCMs are run at a very coarse resolution due to the time and cost required to produce simulations at a finer scale. This approach is particularly problematic for accurately simulating cloud processes, because they are so dynamic in both space and time. To improve model forecasts, ASR scientists developed the Multiscale Modeling Framework, which embeds finer resolution cloud models into the GCM, replacing the complex equations formerly used to represent clouds (Khairoutdinov et al. 2001, 2005; Randall et al. 2003; Ovtchinnikov et al. 2006). This breakthrough nested-model approach, specific to clouds, was shown to successfully transfer the small-scale variability of cloud properties into the large-scale GCMs, and has made significant improvements in the representation of the seasonal and diurnal cycle (Khairoutdinov et al. 2008; Zhang et al. 2008).



To address small-scale atmospheric processes, the Multi-scale Modeling Framework, or MMF, embeds a smaller cloud resolving model into each column of the larger global climate model.

Improved representation of deep convection in models. GCMs have a difficult time simulating deep convection, especially over the tropics. Representation of cumulus convection is an essential part of the feedback mechanisms that modulate climate. Comparing convection schemes against observations is difficult, partly because convection parameterizations are formulated in terms of parameters that have not been adequately observed, and partly because the schemes lack information about parameters that are more directly observable.

The frequency of deep convection predicted by the NCAR single-column model (SCM) at field sites has been far greater than that observed (Klein and Del Genio 2006). A number of ARM field campaigns have been devoted to observing deep convection, and the resultant data have provided new insights into how deep convection behaves. By analyzing years of observational data from the ARM Data Archive, scientists were able to deduce a new convective trigger and closure parameterization, one that determines how much precipitation occurs in a given convective event (Xie and Zhang 2000; Xie et al. 2004). Noticeable improvements in climate forecasts resulted from the new parameterization. Zhang and Wang (2006) demonstrated that the new parameterization addresses the “double” Intertropical Convergence Zone problem. This modified convective trigger was implemented in the NCAR SCM (which is essentially an isolated column of a global climate model) and was shown to yield an improved simulation. The convective trigger has also been implemented into a Japanese global weather model (Xie and Zhang 2000).

Quantification of land-surface cloud feedbacks. In June 2007, the Cloud Land Surface Interaction Campaign (CLASIC) took place, centered on the SGP site. The primary focus of the study was to evaluate how land surface processes influence the evolution of cumulus convection, especially the stages leading from cumulus humilis (fair weather clouds) to cumulus congestus (storm clouds), and to ascertain the relative contribution that local water makes to regional precipitation, versus the contribution of water vapor advected into the region. Cumulus convection is an important component in the atmospheric radiation budget and hydrologic cycle, particularly in the Southern Great Plains during the summertime growing season. Land surface changes associated with plowing, crop rotation, and irrigation can induce changes in the surface latent heat flux, sensible heat flux, albedo, and carbon flux. Changes in surface energy balance and moisture transport to the boundary layer influence cloud processes, thus creating a feedback loop.

Sponsored by DOE in cooperation with other agencies and several universities, researchers used aircraft, satellite, and enhanced surface-based instrument platforms to obtain simultaneous ground and airborne data. Measurements included those related to important hydrologic components in land-atmosphere interactions: soil moisture (the storage reservoir), evapotranspiration (the moisture supply to the atmosphere), and precipitation (the moisture supply to the ground). During the campaign, an increasing and high amount of soil moisture over most of the CLASIC domain was observed, supporting the hypothesis of moisture recycling during CLASIC. Knowledge gained from this study is being applied to improved prediction tools that will benefit a broad spectrum of applications in agriculture ranging from more accurate weather forecasting to improved water management decisions and crop yield estimation.

Modeling regional surface energy, CO₂, and isotope exchanges. Predicting surface-to-atmosphere fluxes of latent and sensible heat, shortwave and longwave radiation, and tracers (e.g., CO₂) is critical to force and test atmospheric models. However, characterizing these exchanges in heterogeneous landscapes, such as the ARM-SGP site, is notoriously difficult. To address this issue, ASR researchers have developed a spatially explicit land-surface model (Riley et al. 2009) driven by regional-scale climate

data from the distributed Mesonet sampling stations and NEXRAD precipitation measurements. The model has recently been applied to estimate spatially and temporally explicit latent and sensible heat fluxes, characterize the SGP region as a net sink of CO₂, and investigate the influence of spatial scaling of land-surface characteristics on regional flux predictions. The model has also been integrated with a mesoscale climate model (MM5) to study land-surface and atmospheric feedbacks associated with land-use (Cooley et al. 2005). These researchers have also shown that clouds, and resulting changes in humidity and the direct/diffuse fraction of solar radiation, can have large effects on surface tracer and energy exchanges (Still et al. 2009).

Stable isotopes in CO₂ and H₂O are important tracers of land-surface and atmospheric exchanges. ASR researchers have developed, tested, and applied a mechanistic land-surface model of ¹⁸O and ¹³C in CO₂ and H₂O (Aranibar et al. 2006, Lai et al. 2006, McDowell et al. 2008, Riley et al. 2002, Riley et al. 2003, Riley 2005, Still et al. 2009). They have used the model to improve our understanding of isotopic exchanges and our ability to use isotopes in testing land-surface models.

Developing a unified model for autoconversion rate in warm clouds. The autoconversion process is a key process that must be parameterized in atmospheric models of various scales—from large eddy simulation models to cloud resolving models to GCMs. In this process, large cloud droplets collect small ones and become “embryonic” raindrops. Accurate parameterization and physical understanding of this process is especially important for studies of the second aerosol indirect effect, when microphysical changes caused by the first aerosol indirect effect inhibit or slow down precipitation formation, and thereby increase the cloud liquid water path (LWP) and cloud lifetime. A new type of parameterization was developed by coupling the threshold function that describes the onset of the autoconversion process, with expressions for the rate function and critical radius (Liu et al. 2007). Comparison of the old and new parameterizations for increasing relative dispersion in a GCM study shows a stronger dispersion effect, which diminishes the global average first aerosol indirect effect by 42% versus 34% in the old scheme (Rotstayn and Liu, 2009).

Precipitation initiation in low mountainous regions. The Convective and Orographically Induced Precipitation Study (COPS) was conducted in the Black Forest region of Germany during 2007. The focus of the study was to advance the quality of forecasts of orographically induced convective precipitation through 4-dimensional observations and life-cycle modeling. COPS was conducted through an extensive collaboration between research institutions from eight countries (Wulfmeyer et al. 2008). The ARM Facility provided the ARM Mobile Facility (AMF) and its extensive array of instruments to obtain a previously unachieved data set on initiation of convection, as well as cloud and precipitation microphysical properties in a low-mountain region.



The AMF in the Black Forest for COPS.

The data collected during COPS allowed a unique view of the complex atmospheric structure during the formation and development of thunderstorms (Kalthoff et al, 2009). Strong spatial inhomogeneities in the water vapor field (Kneifel et al, 2008) could be observed and validated by aircraft observations. Early results indicate deficiencies of mesoscale models in the simulation of convection initiation and the importance of convection permitting resolution of future operational weather forecast and regional climate models (Arpagaus et al. 2009; Trentmann et al., 2009). Data provided by the AMF during COPS

have been used to evaluate forecast models (Crewell et al. 2008), for data assimilation, and for process studies, particularly focusing on the microphysical properties of clouds. The data will further be analyzed to improve the prediction and quantification of precipitation for enhanced ability to forecast the likelihood of extreme weather events.

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